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SOME OF THE LAWS CONCERNING VOLTAIC CELLS.

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[Considerable difference of opinion exists amongst authorities with regard to the magnitudes known as difference of potential and electromotive force, respecting their relation to each other, and to the phenomena which occur in voltaic cells. Professor Landis has made analytical and experimental investigations in this field the results of which are presented in his paper.]

THE purpose of this paper is to discuss some of the difficulties which have arisen in endeavoring to understand the fundamental phenomena of voltaic cells, and to discuss the solutions of those difficulties which have aided in securing a firmer grasp of that group of phenomena.

It will first be considered what transformations of energy are characteristic of those objects to which the name voltaic or galvanic cell has been given. Ostwald says, "We will give the name cell to any arrangement by means of which electrical energy can be obtained from chemical energy."[1] Le Blanc gives a more comprehensive definition by calling "any arrangement"

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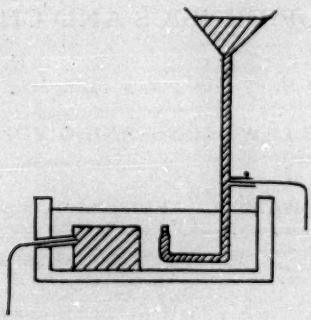
a galvanic cell "which through chemical reaction or physical processes, such as diffusion, etc., is capable of producing electrical energy; whether the reaction takes place between a solid and a liquid, or two liquids, is of no account." [2]

The statement of Ostwald can be found in many places, but the customary application of the name is unquestionably wider than this statement will allow. That of Le Blanc is more inclusive, but how much more is rather uncertain.

It seems advisable and convenient to include amongst voltaic cells series of conductors by which electrical energy can be generated from one or more of these transformations of energy:

FIG. 1.



(1) of chemical energy; (2) of energy of solution; (3) of heat energy *when the series is at uniform temperature*; (4) of energy arising from changes in the volume of the conductors; (5) of kinetic and potential energy.

Des Coudres [3] has made a study of cells in which kinetic and potential energy are transformed into electrical. The simplest of these is shown in Fig. 1. The tube with the funnel contains mercury which cannot flow from the tube because of a fine capillary at the lower end. The larger vessel contains a solution of a mercury salt. The smaller vessel contains mercury. The current flows in the cell from the column to the smaller vessel. This is accompanied by a decrease in potential energy since the height of the column decreases. The E.M.F. of such

a cell is only .0000153 volts for a column of mercury 760 mm. high at 20° C.

It is with hesitation that photo-electric generators have been excluded from a place amongst voltaic cells. These generators have electrodes of the same substance in a solution; as copper oxide in a solution of potassium or sodium hydroxide. They generate electrical energy by the transformation of light energy which is allowed to fall on one of the electrodes. Thermo-electric generators in which heat energy is transformed into electric energy only when differences of temperature exist at different parts of a series of conductors are excluded from amongst voltaic cells even when one of the conductors is an electrolyte.

The most important attribute of a cell is that to which the name E.M.F. is given, but this name is not limited in its use to cells alone; we often speak of E.M.F.'s other than those of cells. It is therefore customary to give a general definition of E.M.F., and afterwards a specific definition of the E.M.F. of a cell. Since this is one of the commonest names in physics, agreement ought to exist in stating what is intended by it; but an examination of some of the best authorities reveals considerable difference of practice in this respect.

I shall endeavor to show that in the class of magnitudes known as differences of potential there exists a smaller class of magnitudes which possess certain characteristics not possessed by the other members of the class in which they are found. To the members of the smaller class convenience and precision join in dictating a more exclusive application of the name E.M.F. E.M.F.'s are therefore differences of potential of a certain variety; every E.M.F. is a difference of potential, but not every difference of potential is an E.M.F. If this task were attempted from the statements of physicists concerning the use of the name the endeavor would appear rather difficult, for in their divergent views, of which at least three exist, there is nothing to suggest a consistency running through the use of this name, a usage which it would seem well to point out and advisable to follow.

One view is that E.M.F.'s are not differences of potential, but causes of differences of potential.[4] It is said that "there must be cause (*i.e.*, a force present) which drives a certain quantity of positive electricity from one metal to another across their surface of contact, and permanently maintains one at a higher

potential than the other. This force is named E.M.F." [5] But some of those who give this definition qualify it by saying that E.M.F. and difference of potential are often used interchangeably. The authority just quoted in distinguishing between them says: "Potential difference and E.M.F. are very frequently used as identical conceptions. Strictly, they are not so, although both of these magnitudes are expressed by the same numbers in the same system of units. We define potential difference as that difference in the electrical state of conductors in consequence of which a quantity of electricity is driven from one to the other. . . . Potential difference is the cause of that flow of electricity which tends to produce equilibrium of electricity and the same potential throughout conductors which are joined. As the E.M.F., on the contrary, we designate that *force* which disturbs the equilibrium and drives electricity from lower to higher potential. The magnitude of this force we measure by means of the potential difference it produces." [6] It seems exceedingly confusing to measure a force by a difference of potential which is not a force. Moreover, if the difference of potential is the thing to which the name E.M.F. is nearly always applied it is very misleading to present another use which is exceptional.

One authority turns this view around and states that difference of potential is the cause of E.M.F. "Just as in water-pipes a difference of level produces a pressure, and the pressure produces a flow as soon as the tap is turned on, so difference of potential produces E.M.F. and E.M.F. sets up a current as soon as a circuit is completed for electricity to flow through. E.M.F. may therefore be conveniently expressed as difference of potential and *vice versa*, but the student must not forget the distinction." [7]

The third view is that E.M.F. and difference of potential are one and the same. Maxwell says: "When two conductors at different potentials are connected by a thin conducting wire the tendency of electricity to flow along the wire is measured by the difference of potential of the two bodies. The difference of potential is therefore called the E.M.F. between them. E.M.F. cannot in all cases be expressed in the form of a difference of potential. These cases are, however, not treated of in electrostatics. We shall consider them when we come to heterogeneous circuits, chemical actions, motions of magnets, inequalities of

temperature, etc." [8] Maxwell is therefore only a partial adherent of this view, but he does not give any characterization of those E.M.F.'s which cannot be expressed as differences of potential. J. J. Thomson says in defining difference of potential that "the electric potential at a point P exceeds that at a point Q by the work done *by the electric field* on a body charged with unit of electricity when the latter passes from P to Q;" [9] and in defining E.M.F. from a point A to a point B that it is the work done by electric forces on a unit charge of electricity in going from the point A to the point B.[10] These definitions agree so closely that it seems fair to take it that J. J. Thomson would use difference of potential and E.M.F. as equivalent terms.

Turning to the definition of the E.M.F. of a cell we find that one of the authorities who adopts the first view [11] says that the E.M.F. of a cell is the sum of the E.M.F.'s at the surfaces of contact of the conductors which constitute the cell. This is an error in that it locates all the components of the E.M.F. of all cells at these surfaces of contact. Maxwell [12] says that the E.M.F. of a cell is the difference of potential between two conductors of the same substance attached to the terminals of the cell. J. J. Thomson [13] gives a proof of this proposition which Maxwell seems to consider as a definition of the E.M.F. of a cell. But he first deduces from the law of the conservation of energy an equation between the chemical energy which disappears in a cell and the heat energy which appears in the circuit in the same time t. The first is CWt in which C is the current, and W is the mechanical equivalent of the chemical energy which disappears per unit of electricity. The second is $C^2Rt + C^2rt + PCt$ in which R is the external, r the internal resistance, and P is the heat in joules which appears or disappears at the junctions of the dissimilar conductors per unit of electricity. These are equal if all the chemical energy which disappears reappears as heat and we have $CWt = C^2Rt + C^2rt + PCt$. This is reduced to $W - P = C(R + r)$ and the quantity on the left-hand side of this equation is defined as the E.M.F. of the cell. It is left to the reader to find the relation between this definition and the general definition already given.

In proceeding from the view that E.M.F.'s and differences of potential are the same, to the view concerning their relation which will be presented in this paper, two other considerations

are intimately involved: physicists have not distinctly set forth criteria by which we can recognize work done when a current flows from one part of a circuit to another; nor do they plainly distinguish between "work done *by* a current" and "work done *upon* a current." Nevertheless both of these questions are of fundamental importance in forming clear and adequate conceptions of the processes occurring during the flow of electricity.

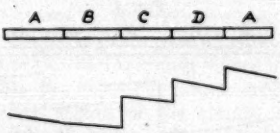
We will first consider one of the most important of those processes of which it is said when they occur that work has been done during the flow of a current. Such a process occurs when a current flows from one part A to another part B of a homogeneous conductor at constant temperature and consists in a generation of heat which is non-reversible in that there is no corresponding destruction of heat when the current is reversed. Only when the resistance of the conductor is constant for all currents are these quantities of heat proportional to the squares of the currents. In such a case it is said that *work is done by the current* in flowing from A to B in overcoming the resistance of the conductor. This work is measured by the heat generated. Further it is said that a fall of potential exists in the conductor from A to B which is measured by the work done per unit of electricity. Changes of potential in conductors of this kind differ from changes of potential in conductors of other kinds in that all of this kind involve a consumption of electrical energy. None of them involve a generation of electrical energy. Nor can we in any way arrange conditions so as to cause them to involve a generation of electricity energy. It would seem that few physicists would maintain that these changes of potential which always involve a consumption of electrical energy are E.M.F.'s. For the sake of convenience we may refer to them as *changes of potential caused by resistance.*

From this simple process which occurs in a homogeneous conductor we may proceed to the processes which occur between a part of one terminal conductor and a part of the other terminal conductor of a type of cell that can be very easily conceived and to which a Daniell cell approximates, but which as far as is known does not exist. These are what may be called *simple chemical cells.* In them the only energy transformations which occur when they produce a current are transformations of chemical energy, and the generation of heat caused by the

resistance of their conductors. Such a cell can have three or more conductors, but we will consider one like the Daniell which consists of five homogeneous conductors. In the Daniell these are copper, zinc, an aqueous solution of zinc sulphate, an aqueous solution of copper sulphate, copper. At three of the junctions of such a cell chemical changes can occur. A chemical cell of this genus can belong to one of many species, but we will only consider several of them.

First we will consider a cell in which when it generates a current chemical energy disappears at all of the three junctions. In this case it would be said that *work is done upon the current* at each of the junctions which is measured by the chemical energy which disappears, and that a rise of potential exists at each of them which is measured by the work done per unit of electricity.* Besides these rises of potential, a fall of potential

FIG. 2.



caused by resistance exists in each of the conductors of the cell. Each of the former involves a generation of electrical energy and in this respect differs from the latter which involves a consumption of electrical energy. Each of these rises of potential is called an E.M.F. and their sum is called the E.M.F. of the cell. The E.M.F. of this cell consists of three components. Fig. 2 is a diagram which represents the changes of potential in this cell assuming that the falls of potential caused by resistance are straight line falls.

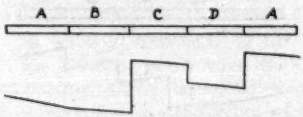
A cell of another species is one in which chemical energy disappears at two of the junctions, say the second and fourth, and appears at the third. Here work is done *upon* the current at junction two and four, and work is done *by* the current at junction three, and a rise of potential exists at each of the former,

* When it is said that a rise of potential exists at a junction it is to be understood that the current arrives there at a lower potential and leaves at a higher.

and a fall of potential at the latter. This fall of potential involves a consumption of electrical energy just as do the falls of potential caused by resistance. But it is far more closely related to the rises of potential which generate electrical energy by the consumption of chemical energy both as regards the processes involved and the laws governing those processes. In view of this relationship and for the sake of convenience, this fall of potential and others like it are called E.M.F.'s as well as those rises of potential to which they are related. And the E.M.F. of the cell is the sum of the rises of potential minus the related fall of potential. Fig. 3 is a diagram which represents the changes of potential in this cell.

These conventions laid down regarding the two simple chemical cells will apply to all of them, but some difficulties arise when what may be called *complex chemical* cells are considered. One genus of these comprises cells in which two chemical processes

FIG. 3.



occur at the same junction; one of which involves a disappearance, the other an appearance of chemical energy. Two modes of interpretation are possible; either that each change of energy contributes its own change of potential, or that all or part of the energy of one process is converted directly into the energy of the other. It is unnecessary to discuss these two modes of interpretation here. Another genus of complex chemical cells comprises cells in which, besides the generation of heat caused by resistance, there are not only changes of chemical energy, but also changes in other kinds of energy which, however, exactly counterbalance.

Therefore in every chemical cell producing a current there occurs besides the generation of heat caused by resistance, a decrease in chemical energy; if any other changes of energy occur, they counterbalance. And the disappearance of this chemical energy with the flow is our fundamental ground for saying that work has been done upon the current, and that an E.M.F.

exists in the cell. Further, the work is measured by the energy which disappears. And the E.M.F. of these cells when producing a current is the sum of all the portions of chemical work done upon the current minus the sum of all the portions of chemical work done by the current per unit of electricity.

The question may well be raised whether these *conventions* regarding work done during the flow of a current are purely arbitrary. Is the law of the conservation of energy involved? Unquestionably the formulation of that doctrine was the chief cause in the development of greater clearness in the conception of the significance of the term E.M.F., and it is very likely if that law did not hold, if a portion of the chemical energy which disappears in a cell did not appear elsewhere, in any form whatever, that it would be said that the work done upon the current is measured by that fraction only of the energy which does reappear elsewhere. Another convention which has been proposed in regard to some of the energy changes which occur in a current bearing circuit is that a rise of potential does not exist in some cases at a region of the circuit where a portion of energy disappears, but that the electricity unites with this energy and carries it to another region of the circuit, disuniting with it there without a fall of potential.[14] Unless such changes in terminology can be shown to correspond to radical differences in phenomena and are appropriately applied to them they cannot be looked upon as aiding in the advance of science.

We are now ready to consider that important law which in different forms is known as Thomson's law. It was first definitely formulated and a mode of deduction given by Kelvin in 1851 following the prior consideration of it by Joule and Helmholtz. As stated by Kelvin it is that "the intensity of an electrochemical apparatus is, in absolute measure, equal to the mechanical equivalent of as much chemical action as goes on with a current of unit strength during unit time."[15] This proposition is erroneous, but the propositions which have been put in its place are not very much better. Nernst says: "It would follow from the law of the conservation of energy, if a galvanic cell neither rose nor fell in temperature when yielding a current, that is, if it neither gave heat to its surroundings nor withdrew heat from them"[16] that its E.M.F. is equal to the decrease in chemical energy per unit of electricity. What he

intends is that if in a galvanic cell the heat generated by resistance be excluded from consideration, and if the sum of the other portions of heat generated is equal to the sum of the portions of heat destroyed, then the remainder of the proposition is true. But even this statement is erroneous unless we exclude from amongst galvanic cells all those cells which involve the generation of electrical energy by transformation of any other forms of energy besides heat and chemical energy. Nernst does not speak of this restriction, and in the absence of any good reasons we may conclude that the restriction is unnecessary. Another statement is that if in a cell all the chemical energy is transformed into electrical,[17] then Thomson's law holds. But this too is erroneous since all the chemical energy and some other form besides could be transformed into electrical energy.

The correct form of this theorem is,—*if in a voltaic cell, the only transformations of energy caused by a current, besides the generation of heat in overcoming resistance, are transformations of chemical energy, or if there are other transformations they counterbalance; then the E.M.F. of the cell during the flow is the net change of chemical energy per unit of electricity which flows through the cell.* No complicated deduction of this theorem is necessary, it follows immediately from the conventions regarding the E.M.F. of a cell which have already been laid down.

So far we have only considered criteria for the existence of changes of potential and the modes of measuring them in a cell which is bearing a current. Not one word has been said regarding a cell which is not bearing a current. Those phenomena caused by a current, those disappearances and appearances of certain forms of energy which are the only grounds so far given for asserting the existence of changes of potential, and are the direct means of measuring them: all those phenomena cease when the current ceases. Nevertheless we agree to say that a change of potential exists from one terminal of a cell to the other even when no current flows through it, and we agree upon a mode of measuring it based upon phenomena which occur when a current does flow through it. Suppose that a steady current is flowing through a homogeneous conductor MN of the same substance as the terminal conductors of a cell C, and suppose that the positive terminal of the cell is joined to a part P of the conductor MN while the negative terminal is joined to a

part Q of the conductor which is at a lower potential than P. The fall of potential in MN can always be made so great that, depending on the positions of P and Q, a current flows through the cell in the ordinary direction, or no current flows, or current flows through the cell in a direction opposite to the ordinary. Suppose the first of these cases exists. We can by altering the positions of P and Q cause the current in the cell to diminish. From the law of the conservation of energy and Ohm's law it can be deduced that in such a case each fall of potential from P to Q in the conductor MN is more nearly equal to the E.M.F. of the cell than any which precede and that they can be made as nearly equal to the E.M.F. as we please by making the current small enough. Therefore, when there is no current in the cell we say that a rise of potential which is an E.M.F. exists in the cell from the negative to the positive terminal as when there is a current; and we can say that this E.M.F. is equal to the fall of potential from the part P to the part Q of the conductor MN to which the terminals of the cell are joined when no current flows through the cell.

We will now briefly recapitulate the views which have been presented concerning work done during the flow of a current and changes of potential in conductors.

First: Excepting those processes which are not caused by the current, such as local action in a cell; and those processes in which a portion of energy disappears at a part of a circuit and an equal portion of energy appears at the same part of the circuit;—excepting these cases, whenever energy not electrical appears or disappears at a part of a circuit, we say that work has been done *by* the current or work has been done *upon* the current. This performance of work is our fundamental ground for asserting a change of potential at that part of the circuit at which it occurs. Further, the work performed is measured by the energy which appears or disappears, and the change of potential by the work done per unit of electricity.

Second: There are changes of potential caused by currents flowing against resistance. All of these involve a consumption of electrical energy and we cannot in any way arrange conditions so as to cause them to involve a generation of electrical energy. These are not E.M.F.'s.

Third: There are changes of potential when a current is

flowing, not caused by resistance, but arising from other causes. These involve either a generation or a consumption of electrical energy. Those which involve a generation of electrical energy are E.M.F.'s. Those which involve a consumption of electrical energy, although in this respect resembling changes of potential caused by resistance, are not classed with them, but are called E.M.F.'s too, because of their relationship to changes of potential which involve the generation of electrical energy, both in regard to the processes involved and in regard to the laws governing those processes.

Finally: There are changes of potential when no current flows. These are not caused by resistance, and are called E.M.F.'s too, because of their relationship to the preceding changes of potential.

Before applying the general principles which have been laid down to particular cases it will be advantageous to consider another type of voltaic cell besides the chemical cell. These are what may be called heat cells. In them the only transformations of energy which occur when they generate a current, besides the generation of heat caused by resistance, are transformations of heat energy, or if there are other transformations of energy they counterbalance. If in one of these cells the heat caused by resistance is neglected, and if the cell is isolated from all outside sources of energy, it will suffer a decrease in heat energy. If portions of heat equal to the portions which disappear are transmitted to the cell it undergoes a process similar to the isothermal expansion of a gas. The E.M.F. of such a cell when producing a current is the net decrease of heat energy per unit of electricity neglecting the heat caused by resistance.

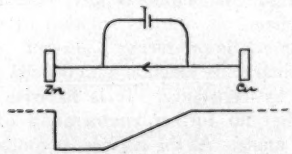
To this class approximate some of the members of that class of cells known as concentration cells. One of these consists of a weak amalgam of zinc, a dilute aqueous solution of zinc sulphate, a weaker amalgam of zinc. The current flows from the stronger to the weaker amalgam in the cell, the concentrations of the amalgams become more nearly equal, the concentration of the solution increases at the anode and decreases at the kathode. Another concentration cell consists of silver, a weak aqueous solution of silver nitrate, a stronger aqueous solution of silver nitrate, silver. The current flows from the weaker to the stronger solution; the silver decreases at the anode and

increases at the kathode; the concentration of the weaker solution increases at the anode and that of the stronger solution decreases at the kathode. That these cells shall be heat cells it is necessary for these changes in concentration to occur without any changes in chemical energy or energy of solution. These cells have E.M.F.'s of only about .05 volts, but they are of much importance in the theories of Nernst regarding cells.

We will now turn to the processes which occur in a Daniell cell consisting of the conductors: copper, zinc, an aqueous solution of zinc sulphate, an aqueous solution of copper sulphate, copper—when the cell is producing a current. From a part of the negative terminal to its junction with the zinc there is a fall of potential caused by resistance. It is likewise with the other conductors, so that no further enumeration of these falls of potential need be made. At the surface of contact of the copper and the zinc two processes may occur: first, there may be caused a very slow union of the two metals with a corresponding decrease of chemical energy; and second, heat may appear or disappear. If the first of these processes occurs, it is so slight as to be negligible. The second is almost so. According to the experiments of Jahn[18] .585 calories disappear per ampere per hour at the surface of contact of the copper and the zinc. Respecting the change of potential which occurs at this junction two views have been put forward which have led to much controversy. It is the opinion of Chwolson, the author of one of the most recent text-books on physics which is gaining well deserved recognition, that this controversy has not yet established either of these two views. One group of authorities amongst whom are Maxwell, Lodge, Ostwald and Nernst, while not always explicity stating it, start from the convention which has already been laid down as the fundamental ground for asserting the existence and as the only means of direct measurement of a change of potential at any part of a circuit conveying a current. This convention for this particular case is that, if during the flow of a current energy disappears at such a junction, work has been done *upon* the current, which is measured by the energy which disappears, and a rise of potential exists there which is measured by the work done per unit of electricity. From this convention and the experiments of Jahn, it follows that a rise of potential of only .00068 volts exists at the junction of

the copper and zinc. Another group of authorities including Kelvin attribute to this junction a rise of potential varying from ½ to 1 volt. Kelvin [19] takes his stand largely upon experiments of Erskine-Murray [20] with discs of copper and zinc. One of these was attached to an insulating support, the other was uninsulated. They were placed facing each other about 1 mm. apart in dry air and joined to an electrometer. If they were also

FIG. 4.



connected by wires of copper to a copper conductor, it was found that a fall of potential of about 0.65 volts must be produced between the points of the copper conductor to which the wires were attached by sending a current through the conductor, in order to obtain no difference of potential between the discs as determined by no deflection of the electrometer when the discs are separated. The changes of potential which Kelvin regarded as indicated by this experiment are shown in Fig. 4, in which

FIG. 5.

the plates have been separated for convenience in representation. Lodge has opposed Kelvin's interpretation and gives instead Fig. 5. The advocates of the second view oppose the convention adopted by the advocates of the first view by maintaining that a theorem is deducible from the second law of thermodynamics which asserts that if Π is the heat in joules generated per unit of electricity at the junction of two metals, e the rise of potential there, and T its absolute temperature; we have $\Pi = T\frac{de}{dT}$. Then only when e is proportional to the absolute temperatures does

$e = \Pi$. These two views cannot be further discussed except to say that Lodge [21] has raised grave doubts concerning the validity of this theorem. A deduction of it can be found in the recent work of Chwolson.[22] Of course it is plain that the second view is contradictory to views of changes of potential in conductors presented in this paper. Turning to surface of contact of the zinc and zinc sulphate solution there is a gram equivalent of zinc transformed from the solid state to the state of zinc in an aqueous solution of zinc sulphate per 96540 coulombs accompanied by an increase in the concentration of the solution. At the surface of contact of the two solutions a part of a gram equivalent of zinc sulphate in aqueous solution replaces an equal part of copper sulphate in aqueous solution which may or may not be accompanied by changes in concentration. At the junction of the copper sulphate solution and the copper a gram equivalent of copper is transformed from the state of copper in an aqueous solution of copper sulphate to the solid state accompanied by a decrease in the concentration of the solution. Finally heat may appear or disappear at each of these junctions. No method has as yet been found for determining the values of the changes of chemical energy and solution energy at these junctions. The values of the portions of heat energy which appear and disappear at these junctions are still uncertain, nor has the total heat in the cell apart from resistance been directly measured. Therefore no direct determination of these component changes of potential can be made. But an approximate value of the resultant rise of potential from the zinc to the copper can be obtained from the resultant decrease of chemical energy which occurs by neglecting all other transformations of energy. For every 96540 coulombs which pass through the cell there is a net decrease of chemical energy of 25,050 calories or 105,000 joules, so that the E.M.F. of a Daniell cell is about 1.09 volts.

The most accurate measurement of the E.M.F. of a cell is, when no current is flowing, by the convention already laid down; or, when a current is flowing, by finding the product of the current and the resistance in the circuit outside the cell. This second method is worth considering. It rests upon the doctrine of the conservation of energy and Ohm's law in its *fundamental* form. As often stated Ohm's law asserts either directly or

indirectly that the current in a conductor or in a circuit is equal to the E.M.F. between the ends of the conductor or in the circuit divided by the resistance of the conductor or the circuit. Sometimes it is required that the current must be steady, but this is often omitted. It is frequently maintained that the important point[23] of the law is that the ratio of the E.M.F. to the current is constant for all currents with the same conductor at the same temperature. But the form which has been given contains not the slightest intimation of this important point; and indeed as far as that form goes the ratio of the E.M.F. to the current could vary and the law still be true. If this ratio did vary the resistance would vary with the current. It is greatly to be deplored that these differences exist in the conceptions of the import of this fundamental law of electricity even if the computations made by it lead to no errors. All forms of Ohm's law either rest upon, or are nothing more than a statement of the convention that the resistance of any conductor, from a part A to a part B, to a current flowing from A to B, is the fall of potential, caused by resistance, divided by the current. It is to be particularly noted that this proposition is a convention, and like all conventions cannot be subjected to proof. It can only be examined from the point of view of convenience in studying phenomena. It leaves open the question whether the resistances of the same conductor at the same temperature are or are not equal for all currents. Experiments of the highest accuracy enable us to answer the question affirmatively for solid conductors, non-electrolytic liquid conductors, and electrolytes which are not parts of cells for all currents having the same distribution in the conductor. From this short digression on Ohm's law we will now return to the measurement of the E.M.F. of a cell by means of the current in its circuit and the resistance external to the cell. Starting from the convention which has just been stated we have for a circuit that the total fall of potential caused by resistance, V_r, divided by the current, I, is equal to the sum of the external and internal resistance of the circuit, $R + r$. Or $R + r = \frac{V_r}{I}$. This equation does not as yet contain the E.M.F. of the cell. But *if the net decrease of energy in the cell, apart from the heat caused by resistance, is the only transformation of energy in the circuit besides the generation of heat caused by the resistance of the entire*

circuit, we have by the law of the conservation of energy that the former is equal to the latter, and the E.M.F. of the cell E is equal to the fall of potential caused by resistance. This gives $R + r = \frac{E}{I}$ from which E can be obtained by making R so large that r is negligible and measuring I and R. The equation $R + r = \frac{E}{I}$ with the accompanying restriction constitutes the form of Ohm's law involving only the E.M.F. of the generator.

One of the difficulties which arises with voltaic cells is in regard to the manner in which Ohm's law applies from one terminal to the other of a cell through the cell. In the general equation for Ohm's law $C = \frac{E}{R}$ what does E represent when the equation is applied to this particular case? The customary definitions of E.M.F., with the usual statements of Ohm's law, give not the slightest clue as to what E represents. This difficulty is overcome immediately by means of the fundamental form of this law which has been put forward. A direct application of this form of the law to the particular case under consideration would be to measure the heat caused by resistance in the cell per unit of electricity which is the fundamental mode of measuring a fall of potential caused by resistance and then to find the ratio of this fall of potential to the current. This ratio is the resistance of the cell. This would be an application of Ohm's law to the cell. The experimental difficulties of this method of measuring the resistance of a cell are well nigh insurmountable; and, as far as I know, no attempt has ever been made to overcome them. Therefore, some indirect method must be established.

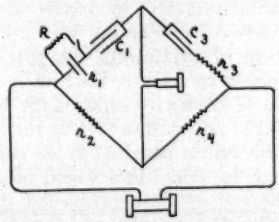
One of these which was much in vogue until about a decade ago, and has not yet been relegated to its proper place in all text-books is Mance's method. Two conditions must be satisfied in this method: the E.M.F. of the cell and its resistance must be constant for two widely differing currents.

A far better method is to measure the rise of potential through the cell from the negative to the positive terminal when a current, which can be found, is flowing through it; and immediately after opening the circuit, before disturbing changes in the cell can occur, to measure the rise of potential through the cell when no current is flowing. The first rise of potential will be denoted by e; the second rise of potential which is the E.M.F. of the cell with no current will be denoted by E; the

current will be denoted by I. The rise of potential through a cell when a current is flowing is less than the E.M.F. of the cell with that current by a magnitude which is the fall of potential caused by resistance. *Hence if the E.M.F. of the cell with a current is equal to its E.M.F. immediately upon the cessation of the current,* $E - e$ is the fall of potential in the cell caused by resistance, and the ratio of this to the current flowing through it, is by the convention already stated *the resistance of the cell.* If r is the resistance of the cell, we have $r = \frac{E-e}{I}$.

Carhart [24] has developed a particular mode of carrying out this general method. He measured the rises of potential in the cell by charging a condenser attached to the terminals of the cell, and discharging it through a galvanometer. By this method

FIG. 6.



Carhart found that the resistance of a Daniell cell decreased from 5.56 ohms with a current of .024 amperes to 4.80 ohms with a current of .190 amperes. It can be urged against these experiments that the E.M.F. with no current which was measured before, instead of after, measuring the rise of potential in the cell with a current, was too great and that this error was greater for the smaller currents.

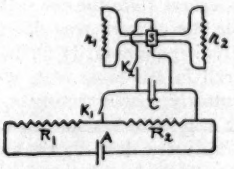
Ayres [25] has devised a modification of the method of Kohlrausch. A Wheatstone bridge is used with the cell and a condenser for one arm, a condenser and adjustable resistance for another arm, and two adjustable resistances forming the remaining arms. The junction of the first two arms is joined to the junction of the last two arms by a conductor attached to a telephone. The current for the bridge is supplied by an induction coil joined to the other pair of junctions. This arrangement permits the measurement of the resistance of the cell through a wide range of current. The current

from the cell flows through an auxiliary conductor attached to its terminals. A diagram of the bridge is shown in Fig. 6. The limits of this paper will not permit a discussion of the theory of this bridge. The equations which hold when there is no current in the telephone are

$$\frac{\frac{r_1 R}{r_1 + R}}{r_2} = \frac{r_3}{r_4} = \frac{C_2}{C_1}.$$

The deduction requires that the E.M.F. of the cell and the resistance are constant for the slight currents added by induction coil to the current of the cell. Ayres found that the resistance of a Daniell cell did not vary more than a few thousandths of an ohm from 1.600 ohms, for currents from a few thousandths to about one-fifth of an ampere. These results are to be expected in view of the experiments made with electrolytes which are not parts of cells.

FIG. 7.



I have performed a series of experiments with a Daniell cell to determine whether the disagreement between results of Carhart and of Ayres arise from errors in the experiments of Carhart or whether they demand another explanation. The method of these experiments differs from that of Carhart merely in the manner of measuring the rises of potential in the cell with and without a current. Instead of a condenser an auxiliary circuit was used. The rises of potential were measured by balancing against a fall of potential in that circuit caused by resistance.

Fig. 7 is a diagram of the arrangement of the apparatus. The rises of potential in the Daniell cell C were measured by the fall of potential in a resistance R_2 in which a steady current was flowing from a storage cell A. This current was maintained steady by keeping the temperature of the storage cell constant and keeping the sum of the resistances R_1 and R_2 in

two boxes always equal to that of one of them with all the plugs removed. The resistance in each of these boxes was of course varied for each adjustment. The current through R_1 and R_2 was measured before and after a series of measurements by balancing a Clark cell across R_2. The Daniell cell had a cylindrical sheet copper electrode 11 cm. high and 8 cm. in diameter which rested upon the bottom of the containing vessel. A saturated solution of copper sulphate was 3½cm. deep in this vessel. A porous cup was placed centrally inside the copper cylinder and a zinc rod 1 cm. in diameter was placed centrally with its lower end upon the bottom of the cup. A saturated solution of zinc sulphate in the cup had its level in coincidence with the level of the solution outside. The rise of potential in the Daniell cell was first measured before any current had been taken from it, except a trace in adjusting the resistance R_2 to balance the E.M.F. of the cell. Then a circuit was closed containing the cell and the box r_1 with a resistance of 15 ohms in it. When the cell had reached a *constant state* the rises of potential in the cell were measured while the current was flowing and immediately after opening the circuit momentarily at the key K_2. The rises of potential of the cell in *the same state* were measured immediately after momentarily disconnecting r_1 from the cell and connecting r_2 to the cell with resistances of 60 ohms and 200 ohms successively. This was done by the switch S. A d'Arsonval galvanometer, not shown in the diagram, was used in obtaining the balances. At the beginning and end of one set of experiments the Clark cell with an E.M.F. of 1.4413 volts was balanced by a resistance of 7957.5 ohms in R_2, showing a remarkable constancy of the storage cell. The current in R_2 was .00018112 amperes. The E.M.F. of the Daniell cell before taking any current from it was 1.0981 volts. The remainder of the results are contained in the two following tables.

Accordingly a slight decrease in the resistance of this cell, beyond the errors of measurement, exist according to this method. This can be explained in another manner which will bring the results of the method of Ayres and this method into agreement. The equation for this method depends upon the assumption that E.M.F. of the cell immediately upon the cessation of a current is equal to the E.M.F. with that current. In the equation for the resistance of the cell $r = \frac{E-e}{I}$, if E, the E.M.F. of the cell immediately upon the cessation of the current,

is smaller than the EM.F. of the cell with the current, the value of the resistance of the cell obtained from the equation will be less than its true value. An increase of the E.M.F. of the cell with the current is therefore all that is necessary to explain the

R_1 when circuit was opened momentarily	E. M. F. of the cell just after opening circuit	r_1 or r_2	
		Resistances in circuit external to the cell	R_1 when the cell was closed through r_1 or r_2
5987.5	1.0844	15.06	2950 2952 2952
5987.5	1.0844	60.06	4752 4752 4752
5987.5	1.0844	200.10	5552 5552 5552

Rise of potential in cell when sending a current through r_1 or r_2	Fall of potential in cell caused by resistance	Current in the cell	Resistances of the cell
.5344	.5500	.03545	15.49
.5347	.5507	.03550	15.51
.5347	.5507	.03550	15.51
.8607	.2237	.01433	15.61
.8607	.2237	.01433	15.61
.8607	.2237	.01433	15.61
1.0056	.0788	.005025	15.68
1.0056	.0788	.005025	15.68
1.0056	.0788	.005025	15.68

contradiction between the results of this method and those of Ayres, and there seem to be no reasons which can be raised with our present knowledge against this explanation. Further experiments with other concentrations of the solutions are desirable accompanied by the simultaneous application of the method of Ayres and this method.

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VALUE OF PIG IRON PRODUCED IN 1908.

STATEMENT BY GEOLOGICAL SURVEY.

The approximate value of the output of pig iron in the United States in 1908 (15,936,018 long tons), as reported by the producers to the United States Geological Survey, was $254,321,000 f. o. b. at the furnaces. This was a decrease of $275,637,000, or 52.01 per cent., from the value of 1907, though the production decreased only 38.19 per cent. The average price per long ton was $15.96, as against $20.56 in 1907, but the latter figure was the highest during the last two decades. These figures are taken from a report on the production of iron ores, pig iron, and steel in 1908, published as an advance chapter from "Mineral resources of the United States, calendar year, 1908." This report contains a map showing the distribution of iron ore in the United States, compiled by E. C. Harder, and another showing the location of blast furnaces in the United States, compiled by W. T. Thom.

SECTION OF PHYSICS AND CHEMISTRY

(*Stated Meeting held Thursday, June* 10, 1909.)

A CRITICAL STUDY OF THE NATURAL CHANGES OCCURRING IN FATS AND OILS.*

BY

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(Contribution from the Food Research Laboratory, Bureau of Chemistry, United States Department of Agriculture.)

(*Continued from page 384.*)

The decomposition of fat by certain of these organisms was shown qualitatively upon plates; the plating medium consisted of an emulsion of palm oil in mineral salt solution to which 1.5 per cent. agar had been added. The bacteria colonies and some of the mould colonies formed clear areas about themselves when plated in this medium; these areas were produced by decomposition of the fat, since microscopic examination of the clear spaces showed that within them the droplets of fat had vanished almost completely. Weighing the fat before and after submitting it to the action of the organisms resulted in a loss in weight, hence the fat was decomposed.

The bacteria were named *Bacillus* α and *Bacillus* β by Rahn. *Bacillus* α is a rapidly moving, liquefying rod, a characteristic distinction between it and *Bacillus fluorescens liquefaciens* could not be determined. It possesses vigorous fat-splitting ability as well as oxidizing power. It is able to destroy oleic acid but does not attack the volatile fatty acids to any great extent.

Bacillus β is a rod in which motion could never be detected. It decomposes fats to a lesser degree than *Bacillus* α. It is uncertain whether this bacillus prefers any fatty acids.

The moulds included *Penicillium glaucum*, another *Penicillium* probably *Penicillium luteum*, a snow-white mould and a gray mould. *Penicillium glaucum* produces greenish yellow spots in

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fat; *Penicillium luteum* (?) colors fat yellow; the snow-white mould gives to fat a magnificent red color. The gray mould decomposes it but slightly.

Experimental proof was obtained by Rahn that each of the bacilli and moulds is able to consume pure glycerol, pure stearic acid and pure palmitic acid.

These organisms in pure culture were also permitted to act upon sterile palm oil or butter fat in the presence of sterile mineral salt solution for several weeks. The acidity and iodine number of the fats increased, while the Reichert-Meissl number decreased. The moulds produced a far more extensive hydrolysis than did the bacilli; they also consumed more volatile fatty acids than did the latter, hence gave rise to a greater decrease in the Reichert-Meissl number. The two *Penicillii* possess strong fat-splitting power; they prefer glycerol and lower fatty acids but do not attack oleic acid. The white mould splits the fats but to a lesser extent than do the *Penicillii*, which it resembles in not attacking oleic acid; it exerts its oxidizing power first upon the volatile acids. Like the gray mould, the yeast decomposes fat to only a slight degree. Rahn suggests that the increase in the iodine number of the fat is due to the formation of unsaturated compounds, possibly from the glycerol.

Several investigators have recorded the changes in the constants of butter fat as the butter became rancid. Koettstorfer [23] mentions that the saponification number of the fat from two samples of butter which had been kept at the temperature of the laboratory—one for six weeks, the other for several months—suffered a slight decrease, which he ascribed to liberation and volatilization of a portion of the fatty acids. However, the decrease in saponification number was so slight that Koettstorfer declares this constant may be used for the detection of foreign fats in rancid butter. Spallanzani [24] determined the Reichert-Meissl number of the fat of some seventy butters of known origin and purity. At the same time, he determined the same constant of the fat of two butters (likewise of known origin and purity) which had become rancid and had been washed. The one sample was eight days old, the other twenty days old. The fat of each had a Reichert-Meissl number much lower than even the minimum value obtained on fresh butter. The older butter had the lower value of the two.

Corbetta [25] determined the Reichert-Meissl number of the fat of the same butter in the fresh condition and at intervals during a period of six months, as the butter became rancid. This constant showed a slight, but progressive, decrease. Washing the rancid butter with water or with sodium bicarbonate solution failed to remove volatile fatty acids, since both the washed and unwashed butter possessed the same Reichert-Meissl number. The butter had been kept in a paper-covered container. On the other hand, von Raumer [26] found that the Reichert-Meissl number of butter fat gradually increased with age and the development of acidity and rancidity. His sample, which was stored in a dark place, was analyzed at intervals during a period of three and a half years. Henriques [27] states that the mean molecular weight of those fatty acids which are readily volatile in steam and are soluble in water, is considerably lower in rancid butter than in fresh butter.

De Kruyff [28] discovered eight new "lipobacteria," which are of very common occurrence in nature, since they were isolated from river water, sewage, excrement of animals and various other sources. However, they occur in such small numbers that de Kruyff was obliged to resort to an elective culture method in order to isolate them. The lipobacteria are aerobes; they first secrete a lipase which produces hydrolysis of the fat; then the organisms oxidize the free fatty acid directly to carbon dioxide and water without the formation of intermediate compounds. Each of the new organisms possesses the power both to split fats and to oxidize the free fatty acids; not one of the bacteria was found to have one and not the other of these properties. Data are given by de Kruyff concerning the hydrolysis and oxidation of triolein, tripalmitin, tristearin and tributyrin, and concerning the oxidation of oleic, palmitic and stearic acids by the lipobacteria. These organisms also destroy elaidin.

The lipase, which these bacteria secrete, is soluble in water, and is able to diffuse in gelatine. A small quantity of acid or of alkali exerts no influence upon the enzyme. When these bacteria were grown upon gelatine plates, in which a glyceride was suspended, each colony formed a transparent circle, due to the diffusion of the lipase secreted by the organisms. The lipobacteria are able to use either organic nitrogen (peptone) or inorganic nitrogen (ammonium chloride or potassium nitrate) as food.

If both denitrifying bacteria and lipobacteria were present, an interesting phenomenon was noticed when the gelatine plating medium contained triolein and potassium nitrate. Microscopic crystals were formed about the colonies. The denitrifiers reduced the nitrate to nitrite, and the nitrous acid caused the rearrangement of the olein into its geometric isomeride, solid elaidin, which formed the crystals.

Bacillus fluorescens is, to a certain extent, a lipobacterium. It secretes the lipase, though but slightly. When the medium contained potassium nitrate and oleic acid, this bacillus caused the oil drops to be filled with crystals. However, when triolein was substituted for oleic acid, crystals formed in both the oil drops and the intermediate spaces. When the medium contained ammonium chloride instead of potassium nitrate, no crystals appeared. De Kruyff states that anaerobes neither hydrolyze nor oxidize fats.

Droste [29] studied the effect of the moulds of the air upon olive oil. A petri dish was covered with agar, which was then exposed to the air. Olive oil was poured upon the culture thus obtained; the dish and its contents were agitated several times daily in order to aerate; they were stored at room temperature in a dark, not very dry place. After the moulds had acted upon the olive oil for a period of twenty days, the oil was without odor, a musty but not harsh taste had developed, the color was unaltered and the oil had become less viscous. It was filtered and analyzed. A great increase in the free fatty acids had occurred; the water-soluble free acids and the Reichert-Meissl number increased slightly. The saponification number and the iodine number also showed an increase. The refractive index remained unchanged, and the response of the oil to the elaidin test was affected to only a slight extent.

Recently Huss [30] has isolated a new fat-splitting organism, *Bactridium lipolyticum*, from milk. It gives a faulty flavor to milk and butter and especially to cream. The bactridium is a facultative aerobe, with flagellæ; it liquefies gelatine. Four enzymes are secreted by the organism, a rennin-like enzyme, a proteolytic enzyme, an enzyme which ferments sugars, with the exception of lactose, and a lipolytic enzyme.

The lipase is able to diffuse through agar and to decompose a layer of beef-tallow beneath the agar. The tallow is changed both macroscopically and microscopically. Macroscopically the

saponified fat may be distinguished from the unchanged fat by a quite white, nearly opaque appearance. It is brittle and moist, and clings to the agar if the latter be lifted. The unattacked tallow, on the other hand, clings to the glass (petri dish). These changes occur after the agar plates have been kept at 37° C. for twenty hours. Under the microscope the particles of unaltered fat have a pale yellow color and no pronounced form. But the particles of decomposed fat have a more brownish tinge, and are usually starlike. Between these small brownish star-shaped particles, long needle crystals, which form clusters, occur abundantly.

When a pure culture of *Bactridium lipolyticum* was added to either sterile or pasteurized cream, which then was kept at 37° C, the cream became rancid in odor and taste at the end of forty-eight hours. The organism attacks both fat and protein. It thrives well in the company of acid-formers up to a certain degree of acid-formation, then is either killed or retarded by the increasing quantity of lactic acid. If the bactridium be accompanied by a number of acid-formers insufficient either to kill it or to retard its growth, the lactic acid prevents either the secretion or the action of the proteolytic enzyme, but fails to influence the lipolytic enzyme; in this case, the cream tastes and smells sourish rancid but not bitter. When unaccompanied by other organisms, *Bactridium lipolyticum* decomposes both butter fat and protein, and ruins the taste of the cream, making it rancid and bitter.

By virtue of its sugar-fermenting enzyme, the bactridium is able to ferment sugars with the exception of lactose; acid is formed in the fermentation. Glycerol is also fermented by the organism with the formation of acid.

Deleamo [31] has investigated the lipase of the moulds, *Penicillium glaucum, Aspergillus flavus* and *Aspergillus niger*. Each of these moulds in the powdered condition possesses the power to split ethyl butyrate, monobutyrin and tributyrin. Ten grammes of each mould were extracted for two days at 37° with a solution which contained 1 per cent. monosodium phosphate and 0.5 per cent. phenol, then filtered. Each extract was permitted to act upon each of the esters. The extract from *Penicillium glaucum* failed to split ethyl butyrate, monobutyrin or tributyrin. The extract of *Aspergillus flavus* had no action on monobutyrin, but produced a weak hydrolysis of ethyl butyrate and tributyrin.

The solution derived from *Aspergillus niger* did not hydrolyze ethyl butyrate but split monobutyrin slightly and tributyrin to a considerable extent.

Organisms, are, in all probability, unable to live upon absolutely pure and sweet fats and oils and to decompose them. However, if the fats be contaminated by even slight quantities of compounds, either organic or inorganic, which supply nutriment to the organisms, the latter thrive and produce changes in the composition of the fats. Apparently organisms are able to live in rancid fats.

Bacteria which decompose fats may be divided into three classes. The first class simply produces hydrolysis of the glycerides. The second class first splits the fats, then consumes the lower fatty acids. The third class hydrolyzes the fats, then consumes the liberated fatty acids of both high and low molecular weight. These decompositions of the fat produce changes in the values of the fat-constants. The first class of bacteria increases the acid value; the second class increases the acid value, iodine number and probably the Hehner number, and decreases the saponification and Reichert-Meissl numbers; the changes in the values of the constants produced by the third class have not been studied. The chief change must be the increase of the acidity. The bacteria which decompose fat are always aerobes and are usually liquefiers; non-liquefiers ordinarily lack the power of attacking fats.

The moulds usually split the glycerides, then consume the volatile fatty acids. As a result, the iodine number, the acid value and probably the Hehner number rise while the saponification number and the Reichert-Meissl number decrease. The bacteria and most of the moulds consume only free glycerol. However, certain moulds possess the power of attacking the combined glycerol of fats, and compounds of etherial odor are thus produced.

The yeasts may be divided into three classes, those which produce hydrolysis of the glycerides, those which attack the combined glycerol with the production of an odor of esters and those which do not have any action upon fats. It has been proved that many fat-splitting organisms secrete enzymes which produce the hydrolysis; possibly all lipolytic organisms act by virtue of lipases.

CHANGES PRODUCED IN FATS AND OILS BY ORGANISMS.—BACTERIA.

Organisms	Fat or Oil	Acidity	Saponification Number	Hehner Number	Iodine Number	Reichert-Meisl Number	Index of Refraction	Lipase	Ester Odor	Investigator	Number
Bacillus fluorescens liquefaciens	Butter fat	+	..	..	..	..	..	..	..	Reinmann	14
Streptothrix alba	Butter fat	+	..	..	..	..	..	..	..	Reinmann	14
Bacillus fluorescens liquefaciens	Butter fat	+	..	..	..	..	..	+	..	Jensen	15
Bacillus prodigiosus	Butter fat	+	..	..	..	..	..	..	..	Jensen	15
Bacillus microbutyricus liquefaciens	Butter fat	+	..	..	..	..	..	..	..	Jensen	15
Streptothrix chromogena	Butter fat	+	..	..	..	..	..	..	..	Jensen	15
Bacillus 2	Butter fat	+	..	..	..	..	..	..	..	Laxa	19
Bacillus 3	Butter fat	+	..	..	..	..	..	..	..	Laxa	19
Bacillus fluorescens liquefaciens	Butter fat	+	−	+	+	−	−	..	..	Laxa	19
Bacillus α	Palm oil / Butter fat	+	..	..	+	−	..	..	..	Rahn	22
Bacillus β	Palm oil / Butter fat	+	..	..	..	..	..	..	..	Rahn	22
Lipo-bacteria	Glycerides	+	..	..	..	..	..	+	..	de Kruyff	28
Bacillus fluorescens	Glycerides	+	..	..	..	..	..	+	..	de Kruyff	28
Bactridium lipolyticum	Butter fat	..	..	..	..	..	..	+	..	Huss	30

YEASTS.

"Sprosspilz"	Butter fat	+	..	..	..	..	..	..	..	Reinmann	14
Mycoderma	Butter fat	..	..	..	..	..	..	..	+	Jensen	15
Saccharomyces	Butter fat	+	−	..	−*	−	..	..	..	Laxa	19
Yeast	Palm oil / Butter fat	+	..	..	..	..	..	..	..	Rahn	22

CHANGES PRODUCED IN FATS AND OILS BY ORGANISMS.—MOULDS.

Mould	Fat or Oil	Acidity	Saponification Number	Hehner Number	Iodine Number	Reichert-Meissl Number	Index of Refraction	Lipase	Ester Odor	Investigator	Number
Hypocreales-nectrieæ	Cocoanut oil	+	..	..	..	..	..	+	Amyl butyrate	Biffin	10
Penicillium glaucum	Butter fat	+	..	..	..	..	..	..	.....	Spaeth	6
Aspergillus glaucus	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Aspergillus niger	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Botrytis cinerea	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Eurotium repens	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Mucor mucedo	Butter fat	+	−	..	−*	−	..	..	.....	Hanus and Stocky	13
Mucor racemosus	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Mucor stolonifer	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Penicillium glaucum	Butter fat	+	..	..	..	..	..	..	.....	Hanus and Stocky	13
Verticillium glaucum	Butter fat	+	..	..	..	..	..	+	.....	Jensen	15
Oidium lactis	Butter fat	+	..	..	..	..	..	..	Ethyl butyrate Amyl butyrate	Jensen	15
Cladosporium butyri	Butter fat	+	..	..	..	..	..	..	Amyl butyrate	Jensen	15
Penicillium glaucum	Butter fat	+	−	..	+	−	−	+	.....	Laxa	19
Mucor	Butter fat	+	−	+	+	−	−	..	.....	Laxa	19
Oidium lactis	Butter fat	+	..	..	−*	−	..	+	.....	Laxa	19
Penicillium glaucum	Palm oil	+	..	..	+	−	..	..	.....	Rahn	22
	Butter fat										
Penicillium glaucum	Palm oil	+	..	..	+	−	..	..	.....	Rahn	22
	Butter fat										
Penicillium luteum?	Palm oil	+	..	..	+	−	..	..	.....	Rahn	22
	Butter fat										
Snow-white mould	Palm oil	+	..	..	..	..	..	..	.....	Rahn	22
	Butter fat										
Gray mould	Palm oil	+	..	..	..	..	..	..	.....	Droste	29
	Butter fat										
Air moulds	Olive oil	+	+	..	+	+*	..	..	.....	Deleamo	31
Penicillium glaucum	Ethyl butyrate	+	..	..	..	..	..	..	.....	Deleamo	31
	Butyrins										
Aspergillus flavus	Ethyl butyrate	+	..	..	..	..	..	+	.....	Deleamo	31
	Butyrins										
Aspergillus niger	Ethyl butyrate	+	..	..	..	..	..	+	.....	Deleamo	31
	Butyrins										
Aspergili	Cocoanut oil	+	..	..	..	..	..	..	.....	Walker	49

EXPLANATION OF THE TABLES.

Under "mould" or "organism" is given the name of the organism.

Under "fat or oil" is given the glyceride upon which that organism was permitted to act.

A sign in the columns of fat-constants signifies that the organism caused an increase (+) or a decrease (—) in the value of that constant. The (*) indicates that the increase or decrease was but slight.

A sign + in the column headed "lipase" means that the secretion of a lipase by the organism was demonstrated.

The + in the column headed "ester odor" signifies that the organism produced an odor of volatile esters in decomposing the glycerides; when the odor recalled that of a particular ester, the name of that ester is given.

The columns "investigator" and "number" give the name of the investigator and the number assigned to his paper in the appended bibliography.

TISSUE-ENZYMES AND THEIR RELATION TO THE FATS.

Enzymes, which are able to produce hydrolysis of fats and of other esters, are found in both the animal and vegetable kingdoms. In the plant-world, the seeds of many species contain more or less fat; in the seeds of these species a lipolytic enzyme is usually present. Sigmund [32] studied the lipase of the seeds of summer rape (*Brassica Napus annua*), winter rape (*Brassica Napus oleifera*), *Ricinus communis, Ricinus major,* poppy (*Papaver somniferens*), hemp (*Cannabis sativa*), flax (*Linum usitatissimum*), pumpkin (*Curcurbita pepo*) and maize (*Zea mais*).

The seeds of each species were ground with water and the resulting emulsions were allowed to stand at least for several hours and at most for two days. The free fatty acids were determined at both the beginning and the end of each experiment; they increased in every case.

In order to exclude fermentation and putrefaction during the splitting of the fat, a protein, to which the lipolytic enzyme clung, was prepared from the seeds and was substituted for the latter in the experiments. The mode of preparation of the

enzyme-rich protein may be described briefly. The seeds of rape or of ricinus, both of which proved to be especially suitable, were finely divided and extracted with water or glycerol; the extract was treated with alcohol whereupon the enzyme and proteins were precipitated. The precipitate was collected upon a filter, washed with alcohol, and dried at 30° C. Weighed quantities of this precipitate in the finely divided condition were mixed with water and a weighed quantity (5 or 10 grammes) of an oil, usually olive oil, of known acid content; the mixture was agitated frequently in order to maintain an emulsion. After a period of twenty-four hours had elapsed, the acid content of the oil was determined. Control experiments were made in which pure egg-albumin was substituted for the precipitate in order to allow for any hydrolysis produced by the proteins of the precipitate. This series of experiments was carried out at ordinary temperature. While 0.20 to 0.52 gramme of the precipitate from rape seed liberated from 51 to 96 milligrammes of oleic acid, the same quantities of egg-albumin set free a much smaller amount of oleic acid,—from 8 to 11 milligrammes. During similar experiments made at 30° to 40° C., the oleic acid, which was liberated by the same quantities of precipitate as used in the preceding experiments, was increased in weight by 10 to 25 milligrammes.

Sigmund concluded that the precipitate contains a lipolytic enzyme, the action of which is slow and of slight intensity.

Green,[33] in studying the germination of the castor-oil plant—*Ricinus communis*—devoted much attention to the lipase of the seeds. The germinating seeds were extracted with an aqueous solution which contained 5 per cent. sodium chloride and 0.2 per cent. potassium cyanide. The extract was dialyzed until almost free from sodium chloride, which hinders the action of the lipase, but the content of the extract in potassium cyanide was kept constant in order to prevent putrefaction. Ten c.c. of the dialyzed extract were mixed by stirring with a fluid ounce of castor oil which previously had been made into a thick emulsion with gum. Litmus solution was added; the mixture was made just alkaline to the litmus, and was placed in a dialyzing tube, which was suspended in 200 c.c. of a solution containing 0.6 per cent. sodium chloride and 0.2 per cent. potassium cyanide. The temperature at which the experiment was made was 37° C., the dura-

tion of the experiment was one week. The litmus gradually became purple, then reddish, and glycerol appeared in the dialyzate which remained unchanged in reaction. Therefore, the lipolytic enzmye of the extract had split the oil into free fatty acid and glycerol. In a control experiment in which the extract was boiled before mixing with the oil-emulsion, neither free acid nor free glycerol was formed.

The lipase, according to Green, acts best in neutral solution; its action is hindered by as little as 0.066 per cent. hydrochloric acid and is stopped by 0.133 per cent. of the same acid. Likewise 0.066 per cent. sodium carbonate slightly retards the activity of the enzyme while 0.66 per cent. of this carbonate produces an entire cessation of the hydrolysis.

He states that the enzyme is present in the resting seeds as a zymogen. Such seeds were ground and freed from oil by means of ether. An extract, which was prepared from the residual powder in the usual way, failed to hydrolyze emulsion of castor-oil. However, this extract became active after it had stood for several days, or after it had been heated at 35° for three hours with weak acetic acid, which was then neutralized. Extraction of the powdered, oil-free, resting seeds with weak acetic acid, at incubator temperature, also furnished a lipolytically active extract. The zymogen was converted into an active lipase by the long standing in solution and by the weak acetic acid. The ricinus seeds also contain a proteolytic (trypsin) enzyme and a rennin ferment.

Browne [34] discovered in rice a lipase which splits the rice oil and thus produces a rancid rice-bran. The acid value of the oil derived from a bran, which had been milled six hours, was much higher than the acid value of the oil prepared from another portion of the same raw rice. The acidity increased in unheated bran with far greater rapidity than in heated bran. Browne ascribes the activity of the lipase in the rice bran to the fact that in the bran a far greater surface is exposed than in the rice.

Connstein, Hoyer, and Wartenberg [35] have made an exhaustive study of the lipase of the seed of *Ricinus communis*,—the castor-oil plant. When seeds and a chloral hydrate solution were rubbed together intimately and then permitted to stand, hydrolysis at first was slight; after a day or two, the velocity increased

and after several days the fat of the seed was almost completely split. The hydrolysis was not hindered by the presence of acid; on the contrary the presence of 2 per cent. acetic acid, $\frac{N}{10}$ sulphuric acid, $\frac{N}{20}$ and $\frac{N}{20}$ sodium acid sulphate hastened the splitting of the fat. Experiments with seeds, from which the oil had been removed, showed that neither water nor acids can decompose the oil-free seeds with formation of acid products; further, control experiments on the reagents gave no increase in the acidity; hence the increase in acidity, when seeds containing oil were used, must have been due to the action of a lipolytic enzyme on the oil. The chloral hydrate was used as a disinfectant. Each of the following fats and oils—tallow, bone fat, cotton seed oil, palm oil, palm kernel oil, rape oil, peanut oil, poppy oil, linseed oil, olive oil, castor oil and fish oil—was submitted to the action of the ground seeds and solution of chloral hydrate (1 per cent.) and acid or acid salt. These fats and oils were split extensively, although not with the same ease and intensity; in some the hydrolysis was complete.

All plant seeds do not possess the lipolytic power to the same high degree; the seeds of the *Euphorbiaceæ* and especially of the ricinus varieties were found to surpass all other seeds in producing hydrolysis. Ricinus seeds retained the power to split fats, after their shells had been removed, and the oil had been extracted either by pressure in the cold or by use of a solvent; the lipase is completely indifferent to treatment of the seeds with ether or carbon disulphide. The lipolytic power of the enzyme is weakened by treatment of the seeds with water, salt solutions, glycerol, etc. This is especially true if the solution of the enzyme be evaporated, even at so low a temperature as 30 to 35°. The power is also lessened if the same sample of seed be used several times, if the seed become too hot during grinding, or if the seed be dried for too long a time at 100° C. Both resting and germinating seeds of ricinus were found to be able to hydrolyze fats with approximately the same intensity. Each kind of seed, in the absence of added acid, produces a slow initial rate of hydrolysis, which becomes far more rapid after the lapse of several days; each, likewise, causes a rapid splitting of fats immediately, provided dilute acid has been added.

Energetic splitting of the fat depends upon both the quality and the quantity of the seed employed. When the quantity of

seed is increased, the hydrolysis becomes more energetic, but the intensity of the reaction is by no means proportional to the quantity of seed used; in fact small quantities of seed have a relatively more energetic action than larger quantities.

The fat itself also exerts an influence upon the hydrolysis. The splitting of various commercial oils does not proceed with uniform ease and intensity. Butter fat is very refractory toward the lipase. This is probably due to the esters of lower fatty acids which occur in this fat. Experiments upon triacetin, tributyrin and triolein showed increasing hydrolysis with increasing molecular weight, but an energetic splitting occurred only in the case of the triolein, for the quantity of acid liberated from triacetin was exceedingly slight, while that set free from tributyrin was rather small. In these experiments, also, it was observed that the slow initial rate of hydrolysis became greater after some time had passed or after a dilute acid had been added. By studying the action of the enzyme upon ethyl acetate, isobutyl acetate, amyl acetate, ethyl sulphate, amyl nitrate and benzyl benzoate, it was found that esters of acetic acid and of mineral and aromatic acids are split but slightly. On the other hand, neutral methyl oleate was attacked quite energetically by the lipase, although the liberated methyl alcohol has a harmful effect upon the enzyme.

Various factors influence the hydrolysis. At least threefold the theoretic quantity of water should be present; greater quantities appear to be of little use but usually do not exert a harmful influence. Sulphuric, phosphoric and acetic acids, sodium acid sulphate and possibly many other substances, which have an acid reaction and are soluble in water, cause the hydrolysis to be rapid from the very beginning. The optimum concentration of all the acids investigated lay between $\frac{N}{10}$ and $\frac{N}{3}$. While acids, which are not soluble in water, may be used, far greater quantities of such acids must be applied. The function of the acid is scarcely to render active a zymogen, for if the seeds be treated with acid and the latter be removed, the seeds possess very slight initial lipolytic power in neutral solution and acid must again be added to produce the strong initial hydrolytic power. A good emulsion is essential for hydrolysis; one part of seed is capable of emulsifying about 100 parts of fat. An almost permanent emulsion may be produced by grinding the fat intimately with either the raw or oil-free seed and then mixing with the acidified water.

Emulsification of the seed and subsequent addition of the fat gives rise to a poor emulsion and consequently an incomplete hydrolysis.

The temperature also plays a rôle. The reaction apparently occurs more rapidly and intensively at a moderately elevated temperature than at lower temperatures. The increase in lipolytic power, which develops on standing when no acid has been added to the emulsion, likewise takes place sooner at a moderately higher temperature. Higher temperatures destroy the emulsion of liquid fats, but exert a beneficial effect upon the emulsions of solid fats. It is not prudent to pass above 40°; at 50° the hydrolysis has been retarded, and a temperature of 100° immediately stops the reaction. Time also plays a part. Although the reaction sets in with extraordinary velocity and intensity, yet a certain period of time is required for hydrolysis. The intensity of the reaction decreases gradually; but even after the emulsion has stood for days, a gradual but continuous increase in free acid occurs. The action of the lipase may be retarded or prevented by harmful admixtures such as alcohol, alkalies, soap, formaldehyde, sodium fluoride and mercuric chloride. The enzyme is indifferent to the presence in moderate concentration of many inorganic salts,—for instance, the sulphates of iron, manganese, magnesium, sodium and ammonium, sodium chloride, and ammonium persulphate.

Lüdy [36] noticed that the tissues of the liver and pancreas possess power to produce extensive hydrolysis of neutral and artificial fats, phenyl esters and acid anhydrides. Muscle tissues according to Lüdy split neutral fats to but a slight extent, and do not even attack tribenzoisin, but are able to hydrolyze phenyl esters and acid anhydrides.

Kastle and Loevenhart [37] have found a lipase in the pancreas, kidney, liver, submaxillary gland and gastric and intestinal mucosa of the pig, and in the liver of sheep, beef, duck and chicken. The enzyme is very stable; after a pig pancreas had been in cold storage for seven days it retained 60 per cent. of its power to hydrolyze ethyl butyrate. A dry, fat-free pancreas retained its lipolytic power in almost full strength for seven weeks. Even a putrefying pancreas still possessed power to split the ester to a slight extent. Aqueous extracts of liver and pancreas of the hog do not lose their power for several days,

while glycerol extracts are still active after several weeks. After studying the influence of about twenty of the commoner antiseptics upon lipase, the conclusion was drawn that by far the greater number of them act in a manner harmful to the enzyme; this is especially true of sodium fluoride, hydrofluoric acid and acids in general. A 0.3 per cent. solution of hydrochloric acid permanently destroys the lipase.

In the extraction of the enzyme from the tissues, if the extract be filtered through paper, the filtrate has far less lipolytic power than is possessed by the filtrate obtained by merely straining the extract through linen or cotton cloth. Repeated filtration at ordinary temperatures almost completely removes the enzyme from solution.

Ethyl butyrate, which is somewhat soluble in water, was found to be a reagent admirably suited for use in the study of lipolytic enzymes. The animal varieties of lipase are able not only to hydrolyze this ester but also to synthesize it from butyric acid and alcohol; in other words, the reaction which is produced by animal lipase is a reversible reaction. The synthesized butyric ester, when formed in small quantities, was detected by its odor. When the experiment was carried out on a larger scale, and more ester was formed, the latter was removed from the reaction-mixture by distillation; the first fraction of the distillate contained the ester.

The optimum temperature of the enzyme is 40° C., at 65° to 70° C. the lipase is destroyed. From experiments on the ethyl ester of each of the acids—formic, acetic, propionic and butyric—which form an homologous series, it was discovered that the higher the molecular weight of the acid, the more readily its ethyl ester is hydrolyzed by lipase. On the other hand, the stability of the ethyl ester of a fatty acid toward normal hydrochloric acid increases with the molecular weight of the fatty acid. The velocity of the hydrolysis of ethyl butyrate increases in almost direct proportion to the concentration of the lipase, but the velocity is not proportionate to the active mass of either butyric or acetic ester. At all ordinary concentrations of both the enzyme and the ester, the hydrolysis is incomplete. However, the reaction approaches completion when very small quantities of ester are used, or when the lipase in itself is especially energetic, or when the lipase solution is exceedingly concentrated.

Loevenhart [38] has found lipase in a number of organs of various animals. In the dog it occurs in the pancreas, kidney, lung, brain, adrenal gland, liver, lymph, lymphatic glands and active mammary gland. In the pig it is found in the pancreas, kidney, liver, intestinal mucosa and subcutaneous fat. The enzyme is present in the liver and heart muscle of man and in the blood of the cat. It is also present in somatic muscle and in the spleen. The most notable occurrences of lipase were in the liver, active mammary gland, blood, lymph and intestinal mucosa. Special interest attaches to the fact that lipase was found in considerable quantities wherever synthesis of fat is known to take place as in the active mammary gland and subcutaneous fat. Lipogenesis is defined as the storage and translocation of fat in the body; both phases of lipogenesis may be brought about by lipase which is able to split fats or to form them according to the prevailing conditions.

Lipase solutions were prepared from liver and from pancreas of such strength that the solution derived from each organ hydrolyzed the same quantity of ethyl butyrate in the same period of time—15 minutes. The solution prepared from each organ was permitted to act upon the ester until the limit of hydrolysis was reached in each case. When each reaction had reached its limit, it was found that the liver lipase had split almost twice as much ester as the pancreatic lipase. When lipase was allowed to act upon butyric ester until the reaction had reached its limit, in the presence of large amounts of the enzyme, the quantity of ester hydrolyzed was proportionate to the amount of enzyme present; with smaller amounts of enzyme, the hydrolysis was relatively more energetic than when larger amounts of lipase were employed. The quantity of butyric ester split by lipase, when the reaction had been permitted to reach its limit, was largely independent of the excess of ester which was present.

Kastle [39] describes certain experiments upon which he was able to base a theory concerning the manner in which lipase hydrolyzes esters and glycerides. Although lipase split diethyl succinate, diethyl fumarate, diethyl oxalate, and diethyl phthalate, the enzyme absolutely failed to produce hydrolysis of sodium ethyl succinate, sodium ethyl fumarate, potassium ethyl oxalate, barium ethyl phthalate, potassium ethyl phthalate, barium ethyl sulphate, potassium ethyl sulphate, barium ethyl p-sulphoben-

zoate, barium ethyl p-nitrososulphobenzoate, potassium ethyl p-nitrososulphobenzoate. Lipase retarded the hydrolysis by water of diethyl p-nitrososulphobenzoate, the action of the enzyme upon diesters of sulphuric and p-sulphobenzoic acids has not yet been studied. These salts of ester acids do not act as poisons toward the lipase; for, if the salt of an ester acid, ethyl butyrate and lipase solution be mixed, the butyric ether is hydrolyzed by the lipase. Moreover, when the salt of an ester acid is mixed with lipase solution, and the mixture is permitted to stand for 24 hours, or longer, the lipase retains its power to split any ethyl butyrate which is added at the end of that period. However, lipase produced hydrolysis of ethyl p-sulphaminebenzoate and of ethyl oxamate. In these experiments the animal variety of lipase was used.

Kastle assumes that the lipase enzyme combines with the molecule of the ester and then produces hydrolysis. The salts of ester acids are electrolytes and hence are ionized; the enzyme cannot combine with an ion and, therefore, does not split such compounds.

He states that animal lipase produces hydrolysis of methyl butyrate; ethyl formate, acetate, propionate, butyrate, isobutyrate and benzoate; amyl acetate and butyrate; and diethyl tartrate.

Lewkowitsch [40] tested the lipolytic power of the lipase of pig's liver upon cotton seed oil, but was unable to obtain an hydrolysis exceeding 3 per cent.; possibly his failure was due to the lack of a complete and permanent emulsion.

Kastle, Johnston and Elvove [41] have succeeded in preparing clear solutions of lipase. They obtained the best results by the following procedure. One gramme of fresh hog liver was macerated with coarse white sand or powdered glass, and mixed with 75 c.c. of water which was then heated to 35° C. Five c.c. of $\frac{N}{10}$ butyric acid were added gradually, the solution was made up to 100 c.c., and the heavy precipitate which had formed was removed by filtration through a folded filter. The filtrate was always perfectly clear and was usually tinted slightly yellow, although frequently it was colorless. Clear solutions were also obtained by means of $\frac{N}{100}$ hydrochloric acid.

The precipitate which remained upon the filter contained lipase; treatment of the precipitate with 0.2 per cent. sodium carbonate solution and subsequent filtration produced a clear,

very active solution of lipase. However, when sodium carbonate solutions of lipase are used in studying the hydrolysis of ethyl butyrate—this ester was used during the research—the liberated butyric acid produces a heavy coagulum of protein during the hydrolysis.

During a period of 15 minutes the lipase, which was present in a clear solution prepared by means of hydrochloric acid, had hydrolyzed at the very least twice its own weight of butyric ester. During the same period the lipase, which was contained in a clear solution prepared by means of butyric acid, had split at least thrice its own weight of ethyl butyrate. The latter solution in a period of 89½ hours had produced hydrolysis of 60 times its own weight of butyric ester, and was still active; the liberated fatty acids had been neutralized with standard alkali at intervals during the hydrolysis.

The clear solutions of lipase may be kept for weeks, and even months, without marked decrease in lipolytic power. The small quantity of acid seems to protect the solution from putrefaction; larger quantities of acid destroy the enzyme completely. The clear solutions may be filtered repeatedly through filter paper without loss in activity. However, filtration through a Pasteur Chamberland filter completely removes the lipase from even its clear solutions.

The hydrolysis of ethyl butyrate by lipase was found to be a monomolecular reaction. From experiments made at intervals of 10° C. from 0° to 40° C., the mean ratio of the velocity of hydrolysis at a higher temperature to that at the next lower temperature was found to be 1.69.

The enzyme suffers no permanent alteration while effecting the hydrolysis of an ester, but retains its complete activity regardless of the amount of substance that it has previously hydrolyzed. The quantity of ethyl butyrate split by lipase is, within certain limits, independent of the concentration of the ester; the hydrolysis is slightly greater in more dilute solutions, because in more concentrated solutions of the ester, the liberated acid has a greater concentration and hence inhibits the enzyme to a greater extent than in more dilute solutions. In dilute solutions of butyric ester, the hydrolysis produced by lipase bears a far greater resemblance to the hydrolysis caused by dilute sodium hydrate than to that produced by dilute hydrochloric acid, when enzyme, acid and alkali are all acting under the same conditions.

Magnus [42] prepared lipase solution from liver by the process of Rosell. Liver juice was treated with an excess of saturated solution of uranyl acetate, and sodium phosphate and carbonate were immediately added to precipitate the excess of uranium. After the supernatant liquid had been removed by filtration, the precipitate, which contained protein, lipase, and uranyl compounds, was permitted to stand for 20 hours in contact with 0.9 per cent. sodium chloride solution; the solution was then filtered. The filtrate, which contained lipase, was made neutral to litmus and was preserved beneath toluol. In his experiments, Magnus used absolutely neutral amyl salicylate; the lipase liberated salicylic acid, which was recovered, and identified and determined colorimetrically by means of ferric chloride.

When the lipase solution was subjected to dialysis in order to remove non-colloidal impurities, the solution within the dialyzer lost the power of splitting amyl salicylate, and failed to recover that power when the sodium chloride content was restored to 0.9 per cent. However, the lipolytic activity returned immediately, when several cubic centimetres of boiled liver juice were added to the solution of inactive lipase. Another portion of the lipase solution was dialyzed for a period of four days, during which the water without the dialyzer was changed twice; these dialysates were united and investigated. The dialysis was continued in running water until the dialyzer no longer contained an active lipase, but only the inactive enzyme.

The dialysates, obtained during the period of four days, were evaporated to dryness; the residue was exhausted with absolute alcohol, which was then evaporated. The alcohol-extract was treated with ether; the portion insoluble in ether was dissolved in water and the solution was neutralized. This solution contained a coferment.

Neither the inactive lipase nor the coferment was able to hydrolyze amyl salicylate; however, the ester was split by a mixture of the solutions of the inactive lipase and of the coferment. The coferment is not altered by boiling its solution, but is decomposed by reduction to ash, hence is not an inorganic body which is permanent at a glowing heat.

Loevenhart [43] found that the addition of bile salts to a dialyzed hepatic lipase solution, which had been prepared by the method of Magnus and was inactive toward amyl salicylate,

caused the enzyme to regain its activity toward that ester. However, the dialyzed lipase solution retained its power to hydrolyze ethyl butyrate, and this action was not accelerated by the addition of bile salts. Therefore, the bile salts are really a coferment but only with respect to amyl salicylate; Loevenhart suggests that possibly the function of the bile salts is to render more soluble the amyl salicylate, which is a very insoluble ester.

Loevenhart and Pierce [44] have studied the inhibiting effect of sodium fluoride upon the hydrolysis produced by hepatic and pancreatic lipase. They used ethyl acetate, ethyl butyrate, diacetin, triacetin, and olive oil. The inhibition of the enzyme action produced by the same quantity of fluoride became less as the molecular weight of the fatty acid increased. The alcohol radical apparently plays no part in the inhibition, since hydrolysis of esters of the same acid with different alcohols is inhibited by the same quantity of fluoride to the same extent.

The lipase is not destroyed by sodium fluoride. With the same quantity of lipase, the inhibition of the enzyme-action increases with the concentration of the fluoride; with the same quantity of fluoride, the inhibition decreases with the concentration of the lipase.

Loevenhart and Pierce offer the following explanation of the inhibiting action of the fluoride. The fluoride reacts with an intermediate compound, which is formed in the action of the enzyme upon the ester, and increases the stability of this intermediate compound, thus retarding the hydrolysis.

Amberg and Loevenhart [45] investigated the inhibition produced by sodium fluoride in the hydrolysis by hepatic lipase of the ethyl esters of acetic, propionic, butyric and valeric acids and of the methyl esters of caprylic and lauric acids. They also found that the inhibition of the ester-splitting decreases as the molecular weight of the fatty acid increases. The equilibrium point in the reaction is not shifted by the fluoride; the production of the equilibrium is merely retarded. Fluorides exert an inhibiting influence at dilutions at which other inhibitants of the lipase have lost their inhibiting power. Amberg and Loevenhart have based a method for the detection of fluorides in foods upon the inhibition by these salts of the hydrolysis of butyric ester by hepatic lipase.

Saxl [46] made a study of the splitting of neutral fats, mono-

acetin, monobutyrin, ethyl butyrate and amyl salicylate by the lipase of various organs, fat tissue and muscle tissue of the dog, rabbit and swine. The fine suspension of the organ or tissue in physiological salt solution was ordinarily used, with the application of toluol as an antiseptic. At the end of each experiment, before titration of the liberated acid, the protein was coagulated by heat and removed from the solution by filtration, so that it could not influence the titration. The lipolytic power of the muscles and of the blood-serum was found to be less than that of the other organs. Saxl states that neutral fat, contained in the organs or added to them, undergoes a splitting to only a very slight degree during the post-mortem autolysis, provided bacterial action be excluded. Butter-fat was split to a slight extent by finely divided lung and liver, in from one to three days at body temperature.

Monoacetin, monobutyrin and ethyl butyrate, when added to the finely divided organs, were split by the latter to a very slight but measurable extent, in one hour at body temperature. When the organs were allowed to act upon the esters for a period of 24 to 48 hours, a strong increase in acidity was shown by the titration. This high acidity, however, was due not only to the hydrolysis of the ester, which had been used, but also to the acid formed during the autolysis. The autolysis experiences a considerable rise through the appearance of slight quantities of acid. Amyl salicylate was hydrolysed by nearly all the organs investigated, with the exception of the muscles. The conclusion is reached that none of the previously recommended methods affords a quantitative study of the ester-splitting, since the values obtained either are so slight on the short periods of digestion that they scarcely lie beyond the limit of error, or, on longer periods of digestion, are obscured by the sources of error which are connected with the methods of determination.

Dieterich [47] has made an interesting study of the changes which occur in swine-fat and beef-tallow before they are rendered. When the fat of an animal, be it cattle, sheep or swine, is removed, it must be spread out and permitted to cool in a cool room. If this procedure is not followed, or if the rough fat lies in deep layers, spontaneous heating occurs, which changes the color of the fat and produces an odor of putrefaction. When such a spontaneously heated fat is rendered, the product differs

from the normal fat not only in color and odor but also in consistency; it separates into a liquid portion and a solid, generally granular part. Dieterich removed the fat from freshly slaughtered cattle and swine, rendered a portion immediately and determined upon the product the values of the fat constants. The remainder of the crude fat was stored for periods of 8 and 14 days at room temperature (15° to 17° C.) and at a temperature of 30° to 35° C. At the end of each period, specimens which had been kept at each temperature were rendered, and the fat was used for the determination of the values of the fat constants. Experiments were made upon the kidney fat of cattle, the leaf fat and the bacon fat of swine.

The most striking change during storage was the increase in the acid value; the change was progressive. More free fatty acid formed during the same period of time at 35° than at 15° to 17°. The higher temperature produced the greatest splitting in the case of the cattle tallow. The swine fats gave rise to a greater stench than did the cattle fat. The refractive index of the fats and their fatty acids showed a tendency to decrease as the acidity increased. The specific gravity of the fats remained unchanged, while that of the fatty acids changed but little. The Reichert-Meissl number oscillated; the iodine number and melting and freezing points of the fats and of their fatty acids underwent very slight changes. The conclusion is reached that a fat is exposed to the greatest changes before rendering and not after rendering.

Work similar to that of Dieterich has been done by Pastrovich.[48] He isolated the membranes which envelop raw tallow, scraped off the adhering fat-particles, wiped off any blood by means of moist filter paper, dried the membranes in air, and chopped the dry product to a fine state of division.

The chopped membrane was mixed with cotton seed oil which contained at least 6 per cent. of water—the quantity necessary for hydrolysis. The mixture was kept in well-stoppered flasks, in the dark, at 35° for as long as 24 days; the flasks were shaken daily. The acidity of the oil became greater day by day; a mixture of 10 grammes of membrane and 100 grammes of oil was hydrolyzed to a greater extent—in the same time—than was a mixture of 5 grammes of membrane and 100 grammes of oil. The hydrolysis of the latter mixture was considerably

retarded by the addition of 2 c.c. of normal sulphuric acid; the presence of 1 c.c. of normal potassium hydroxide retarded the splitting for the first few days, then hydrolysis proceeded as usual.

The autohydrolysis of raw tallow from the marrow, pouch, intestines, and kidneys was also studied. Each variety was freed as far as possible from blood, put through a fat-grinder, and permitted to hydrolyze spontaneously. Usually, though not invariably, water was added to the raw tallow. In all the samples, fatty acids were liberated; the velocity of hydrolysis was greater in the olein-rich than in the olein-poor specimens. The addition of water always aided the splitting of the fat; the greatest acceleration of the hydrolysis was produced by the addition of 12 to 18 per cent. of water. The presence of a small quantity of ammonia or of potassium hydroxide hastened the splitting, but larger quantities of these reagents retarded it. Dilute mineral acids also retarded the hydrolysis as did 2 per cent. solution of chloral hydrate, while 2 per cent. solution of mercuric chloride almost stopped the reaction. Certain of these experiments extended over a period of 163 days.

During the decomposition the odor was sourish and not unpleasant, and frequently recalled that of fresh cheese; the color was that of the raw fat, unless ammonia or fixed alkali had been added, when the color became a reddish yellow. At the beginning of the hydrolysis, free ammonia was evolved; the decomposed raw tallows always contained ammonium soaps, hence were rendered with the addition of a few drops of dilute mineral acid to decompose the soaps. Any free mineral acid was washed out of the rendered fat before the free acids of the latter were titrated. When water or alkalies had been added to the raw tallow, the mineral acid liberated formic acid, which had its origin in the decomposition of the proteins of the membranes. The velocity coefficient of the hydrolysis could not be determined; *i.e.*, gave no constant values for the individual periods of time. The reaction itself apparently resembles the saponification of amyl acetate by ammonia.

Pastrovich was unable to isolate an enzyme from the membranes, which enveloped the raw fat, nor from the fat itself. From the fact that 2 per cent. of mercuric chloride almost stopped the hydrolysis, it was concluded that, even if an enzyme were

present, its quantity must be very slight. Therefore, enzyme action was excluded, and organisms were considered as the cause of the hydrolysis. Aerobic microbes decompose the protein of the membranes with liberation of ammonia, which saponifies the fat. The proteins cannot furnish sufficient ammonia to hydrolyze the fat on the scale with which hydrolysis occurs. However, ammonium soaps are exceedingly labile compounds and readily part with their ammonia. Catalyzers—either the organisms themselves or certain decomposition products of the proteins—liberate ammonia from these soaps; the nascent ammonia is more reactive toward fats than is ordinary ammonia, and hence saponifies the fat. The process of catalysis and saponification is repeated indefinitely, and therefore a small quantity of ammonia may produce hydrolysis of a large quantity of neutral fat with the liberation of fatty acids.

The solutions, which remain upon rendering the decomposed fat, are slightly acid and contain the glycerol produced by the hydrolysis. Whether the glycerol is present in the solutions in its entirety, or whether a portion of it is consumed by the microbes or otherwise changed, was not determined.

Walker [49] has investigated the changes in cocoanut oil both before and after its expression from the copra. He states that most of the free acid with the accompanying bad odor and taste is produced in the copra itself before the expression of the oil; for instance, the oil from a sample of copra, which had been cut into fine pieces and exposed to moist air for one month, increased in acidity from 1.5 to 23.3 per cent. free acid as oleic. Moulds, chiefly *Aspergilli*, which act alone or in symbiosis with certain micro-organisms, first hydrolyze the fat then further decompose it within the copra. The conditions most favorable for the growth of moulds are a moderately high, constant temperature and a content of about 9 to 17 per cent. water; commercial copra contains 9 to 12 per cent. water as a rule, hence forms an excellent medium for moulds. A specimen with a water content of 4.76 per cent. contained neither moulds nor bacteria and the acidity of the oil did not increase; however, samples containing from 23 to 50 per cent. of water, while free from moulds, were infested by bacteria which drew their nourishment from the non-fatty constituents of the copra and which liberated but a small quantity of fatty acid from the oil.

While raw cocoanut oil does not form a suitable medium for organisms, yet, if it be contaminated by water and nutrient matter in sufficient quantities, moulds may develop and rapidly attack the oil. When a pure oil, to which 1 per cent. of water and 1 per cent. of "latic" had been added, was exposed to mould action, for one week in an incubator, the free acid as oleic rose from 0.10 per cent. to 8.63 per cent. To prevent rapid deterioration of cocoanut oil, Walker recommends drying of the copra until its water content is not over 5 per cent., and expression of the oil as soon as possible, avoiding long storage in a warm, moist atmosphere.

Lewkowitsch and Macleod [50] made a study of the hydrolysis of fats by the pancreatic enzyme, steapsin. A series of experiments was made upon fresh ox pancreas which had been freed from fat. The minced organ was ground with water in a mortar, and incubated for 20 hours at body temperature, after thymol or 0.1 per cent. mercuric chloride had been added. The extract was then filtered through paper. The hydrolyzing power of the residue, which remained upon the filter, was far greater than that of the solution itself; apparently filter paper retains the enzyme. The following test for steapsin was used: 2 or 3 c.c. of the extract of the organ were shaken with a few drops of filtered butter fat, phenolphthalein was added, and $\frac{N}{10}$ sodium hydroxide was added until the solution assumed a deep red color. The solution was kept at 37° C. for one-half hour, then the standard alkali was added until the deep red color again appeared. A control experiment was made upon the boiled extract. The extract of ox pancreas did not produce hydrolysis of an emulsion of cotton seed oil; its failure to do so was ascribed to the possible action of trypsin upon the steapsin.

Minced pig pancreas was triturated with twice its bulk of water in a mortar, and filtered through muslin. Loss of enzyme during incubation and by filtration through paper was thus avoided. Measured quantities of the extract, thus prepared, were triturated with 100 grammes of cotton seed oil or of lard until a complete emulsion was obtained. The emulsion was poured into bottles which were well corked in order to exclude the growth of moulds. *Penicillium glaucum* developed in only a few of the samples after four weeks had elapsed; most of the specimens remained free from infection during the entire experi-

ment, which extended over a period of two months. The bottles were kept at the ordinary temperature. Whenever the contents of a bottle tended to separate from the emulsified state, a phenomenon which occurred especially during the first few hours of the experiment, the emulsion was restored by shaking. In the experiments with cotton seed oil, after four days as much as 37.1 per cent. free acid as oleic had been liberated; after 11 days the hydrolysis set free as much as 46.4 per cent., and after 56 days the greatest quantity of free acid was 83.8 per cent. and 86.7 per cent.; neither acid nor alkali had been added during these experiments. However, the addition of either dilute acid or dilute alkali was found to be without decisive influence upon the hydrolysis. An outward sign of the hydrolysis of the oil was the hardening of the emulsion by the liberated fatty acids.

Steapsin split lard at a far slower velocity than it split cotton seed oil. Although it was found that steapsin is not capable of reversing the reaction produced by it, yet it was proved that the enzyme is a powerful fat-splitting ferment.

Gillet [51] has found an enzyme in human milk and in the milk of cows, horses and asses. It produces hydrolysis of monobutyrin, but has no action upon monoacetin, diacetin, triacetin and normal neutral fats; therefore, Gillet proposes that this enzyme be called monobutyrinase and not lipase. The enzyme is not capable of dialysis, and is precipitated by alcohol but again dissolved by water.

Euler [52] studied the relation of the enzyme catalase to the fats. He suggests that the catalase of *Boletus scaber* may bear some relation to the not inconsiderable fat content of that fungus. Very pure and strong catalase solutions were prepared from animal fat-tissues—those of swine. In fact, wherever fat is formed or is decomposed, catalase is found in particularly strong concentration. After a mixture of fresh swine fat with an equal volume of water had stood at 40° C. for one hour, it was cooled to 35° C. and filtered. The clear filtrate exerted an exceedingly vigorous hydrolyzing action upon ethyl butyrate. Extracts of certain seeds, which have been proved to contain lipase, *e.g.*, the seeds of the pumpkin and of the castor oil plant, also produced a vigorous decomposition of hydrogen peroxide. The parallelism of the action of plant and animal extracts in splitting fats and esters and in splitting hydrogen peroxide led to the con-

clusion that the property of the decomposing hydrogen peroxide belongs particularly to the lipase extracts.

Loew [53] maintains that the sole function of catalase, not only in tissues which contain little fat but also in those which are rich in fat, is to decompose the hydrogen peroxide which forms during the oxidation of all tissues. Moreover, tissues which are poor in fat may be rich in catalase and yet entirely lack lipolytic power.

Euler [54] admits that tissues which are poor in fat may be rich in catalase. However, he points out that the intermediate formation in many organs—for example, the fat tissues, of hydrogen peroxide or other peroxides, poisonous to the animal system—has never been proved, although such compounds may possibly be formed. Euler maintains his statement that lipolytic extracts are especially rich in catalase.

In the seeds of plants, and in many animal tissues, is found an enzyme—lipase—which is able to produce hydrolysis of fats and esters, and, according to Taylor,[55] of lecithins. In the case of both the animal and vegetable varieties of lipase the velocity of hydrolysis becomes greater as the molecular weight of the ester or glyceride increases. Both varieties act best at a moderately elevated temperature and are destroyed if heated to too high a temperature; both are greatly retarded in activity by certain reagents, for instance, by sodium fluoride. The animal lipase not only can hydrolyze esters but also can synthesize them from acid and alcohol. The synthesis of triolein by means of a vegetable lipase—that of the castor bean—has been accomplished by Taylor.[56] Lipase may produce complete hydrolysis of fats; this is especially true of the vegetable variety of the enzyme. The enzyme steapsin likewise possesses power to split fats. Green [57] considers steapsin, pialyn and lipase to be synonyms for one and the same enzyme. The changes, which occur in fats and oils before rendering or expression, are probably due to the action of organisms, their enzymes and tissue enzymes. If the enzyme catalase plays a rôle in the chemistry of the fats, that rôle is at present but little understood.

THE ACTION OF ATMOSPHERIC GASES UPON FATS AND OILS.

With the exception of nitrogen, the gases of the atmosphere produce changes in oils and fats which are exposed to their in-

fluence. The action of each of the gases,—oxygen, ozone, carbon dioxide, nitrogen, hydrogen and also water vapor—has been studied by various investigators.

Spaeth [58] quotes Scherer [59] who, in 1795, stated that fats, on long standing, by absorption of oxygen from the air, assume a sharp, biting or burning taste, and become rancid.

Berthelot [60] exposed various synthetic glycerides to the action of the atmosphere by storing them in poorly stoppered flasks. Oleins became acid within a few weeks; valeric acid was liberated from the valerins; butyrins were split rapidly and easily; acetins were soon hydrolyzed, and benzoic acid was, in time, set free from the benzoins. Glycerides and esters of oleic acid and of the lower fatty acids were exposed to the action of the air in the dark for two and a half months both with and without the addition of brass filings which served as a catalyzer. The volume of oxygen absorbed by each compound was determined. Neither benzoic ester, monobenzoins nor butyrins absorbed an appreciable amount of oxygen. Acetic ester and diacetins absorbed a slight quantity of oxygen which became somewhat greater in the presence of the brass filings. The absorptive power was possessed to only a slight degree by valerins in a moist atmosphere and was increased but little by the filings. Natural oleins, of all the compounds examined, had the highest absorptive power for oxygen, absorbing six parts of oxygen for every 100 parts of their weight, with an increase to eight parts per 100 upon the addition of the filings or lead oxide. Oleic ester failed to take up as much oxygen as olein; it absorbed but one part in 100, a ratio which became somewhat greater in the presence of the filings.

Certain glycerides and esters, *e.g.*, those of butyric and benzoic acids, became acid without the action of oxygen. Berthelot decides that oxidation depends particularly upon the acid radical of the ester or glyceride. Acid-formation, however, is connected with the entire molecule of the fat or ester, is due to the splitting into free acid and either glycerol or alcohol, and requires the presence of water or of humidity. This liberation of fatty acid by moisture or humidity, though far slower, is perfectly analogous to the hydrolysis of fats and esters upon heating with comparatively large quantities of water. The spontaneous formation of free fatty acids in natural fats is to be ascribed to the

slow action of the humidity. Foreign bodies are not essential for these decompositions but accelerate them.

Kingzett [61] prepared oleic acid from beef fat, mutton dripping and cocoa butter. Five grammes of each sample of the acid were sealed over water with a measured quantity of air during the month of June. All three samples of acid failed to absorb any oxygen during that time.

Duclaux [62] emphasizes the rôle played by atmospheric oxygen in the decomposition of oils and fats. He states that organisms are unable to attack fats directly, even in mixtures of fat, protein and carbohydrate, although they thrive upon such mixtures. However, the atmospheric oxygen, which acts in both sunlight and darkness, gradually converts insoluble fats into a chain of soluble compounds. Cryptogams utilize these soluble products of oxidation as food, and convert them into carbon dioxide and water.

Gröger [63] made an investigation covering a period of four years, concerning the changes in tallow, bone-fat and palm, cocoanut, cotton seed and olive kernel oils. After the acidity of each fat or oil had been determined, it was divided into four portions, one of which was placed in a tightly stoppered bottle. A second portion was emulsified with 4 per cent. of a concentrated albumin solution and was transferred to a tightly stoppered bottle. A third portion was absorbed by means of strips of dry, fat-free filter paper which were then stored in a large glass vessel with free air access. A fourth portion was treated exactly like the third portion, with the single exception that the filter paper had previously been saturated with a 10 per cent. albumin solution and then permitted to dry.

After a lapse of four years a chemical examination of the samples was made. Those from which air had been excluded by tightly fitting stoppers had increased but slightly in acidity, while the portions emulsified with albumin contained more free acid than the stored samples of pure oil.

Each sample of fat or oil was recovered from the strips of filter paper by means of ether; the residue obtained by evaporation of the ether was washed several times with boiling water, and the acidity of the oil or fat was determined. In the case of the fats recovered from the plain filter paper, cotton seed oil, cocoanut oil and tallow had increased in acidity, while bone

fat, palm oil and olive kernel oil had decreased in acidity since their free acids had been oxidized to volatile and soluble acids. The fats recovered from the albumin-impregnated filter paper were changed to a lesser degree, possibly because this filter paper was less porous than the plain variety. The mean molecular weight of the insoluble fatty acids of each of the fats and oils, which had been exposed to the air, had decreased; hence insoluble acids of lesser carbon content had been formed during the oxidation. Soluble organic acids had been removed from the recovered fats and oils by the boiling wash-water; certain of these acids crystallized out as the water cooled; the mother liquor still contained acids, the quantity of which was so slight that they could not be further investigated.

The crystalline acids from each of the fats and oils were converted into their sodium salts. With the solutions of these salts, silver nitrate produced a white, flocculent precipitate; lead acetate a white, pulverulent precipitate; cupric sulphate a bluish-green precipitate, and ferric chloride a flesh-colored flocculent precipitate; barium chloride did not form a precipitate. The acids distilled apparently undecomposed when heated to a temperature above 300°. From these chemical and physical properties, it was concluded that the crystalline acids were azelaic acid. Moreover, sufficient crystalline acids were obtained from palm, cotton seed and olive kernel oils for elementary analysis and the determination of the melting point and the mean molecular weight; the acid from cotton seed oil was proved to consist entirely of azelaic acid; the acids from palm and olive kernel oils were composed chiefly of azelaic acid with the admixture of a slight quantity of suberic acid.

Gröger was unable to detect free glycerol in the fats and oils which had been exposed to the action of the atmosphere. He concludes that the glycerides are hydrolyzed by moisture, the fatty acids and the glycerol are then oxidized simultaneously by the atmospheric oxygen. The fatty acids are thus changed into acids, poorer in carbon and richer in oxygen, belonging partly to the fatty and partly to the oxalic series.

Ritsert [64] sealed in glass tubes samples of moist swine fat and samples of the same fat, which had been dried for three hours at 140°; the seals were made as close as possible to the fat samples, without burning the latter. The tubes were sterilized by repeated

boiling in a water bath; and then were divided into four sets which were kept in direct sunlight, in diffuse daylight, in the dark at 15° and in the dark at 37°. After the lapse of 8, 20, 40 and 60 days, a tube from each set was opened, and its contents were examined bacteriologically, organoleptically and chemically. Both aerobes and anaerobes were practically absent, the taste and smell remained normal and the acidity did not change. The samples kept in direct sunlight for 60 days had assumed a granular appearance and had altered slightly in taste; these changes were possibly due to the separation of the olein from the palmitin and stearin. Samples of butter, protected from the air by sealing in glass tubes, were perfectly fresh after they had been exposed to the action of light for five months. Moist swine fat was sterilized in cotton-stoppered Erlenmeyer flasks, some of which had been painted black. All the flasks were then exposed to the sunlight. At the end of 5, 20, 40 and 60 days, the contents of an ordinary flask and of a blackened flask were examined with respect to acidity, taste and smell, and presence of organisms. Neither aerobes nor anaerobes were found in any of the flasks. The acidity of the fat in the blackened flasks remained constant and its odor and taste were always those of a fresh fat. The fat in the ordinary flasks showed a progressive increase in acidity, and in rancidity as measured by taste and smell. From these experiments it followed that rancidity was produced only by the simultaneous action of air and light, neither of which by itself gave rise to rancidity.

The absorption of various gases by both moist and dry swine fat was also studied. A 100 gramme sample of the fat was placed in a 400 c.c. flask, which was filled with the proper gas and then connected with a vessel of mercury by means of a vertical glass tube at least 75 cm. in height. As the gas was absorbed, the mercury rose in this tube, and thus gave a measure of the absorption. Ordinary flasks were used in studying the influence of oxygen and of carbon dioxide upon both dry and moist fat, and of air, nitrogen and hydrogen upon moist fat. Blackened flasks were used when oxygen, carbon dioxide and air were permitted to act upon moist fat. In the experiments upon dry fat, the gases were also dried. All the flasks were exposed to the light at a window facing the east, for a period of 30 days. The height of the mercury in each vertical tube was read daily at

6 P.M., as was the temperature and barometric pressure. At the end of a month, no hydrogen or nitrogen had been absorbed; and no air, and practically no oxygen, had been absorbed in the blackened flasks.

In descending order the gases were absorbed by the fat as follows:

Oxygen by dry fat in ordinary flask.
Oxygen by moist fat in ordinary flask.
Carbon dioxide by dry fat in ordinary flask.
Carbon dioxide by moist fat in ordinary flask.
Air by moist fat in ordinary flask.
Carbon dioxide by moist fat in blackened flask.

Both the moist and dry fat in the ordinary flasks, which were filled with oxygen, and also the moist fat in the ordinary flask, which was filled with air, had increased in acidity and had become rancid but contained no organisms. The fats which had been exposed to the action of carbon dioxide had increased in acidity, were without odor, and had a tallowy, flat taste. The acidity of the specimens, which had been kept in hydrogen and nitrogen, and in oxygen and air in the blackened flasks, remained constant and rancidity failed to develop. Ritsert concludes that light causes oxygen, which is without action upon fat in the dark, to become active toward fat and to convert fatty acids into oxy acids which give the rancid taste and odor to a rancid fat. Hence, rancidity is a process of direct oxidation by atmospheric oxygen in the presence of light; and moisture is not a necessary factor in the process.

Ballantyne [65] exposed olive, rape, castor, peanut, cotton seed and linseed oils to the air and direct light of the sun for six months in uncorked bottles, which were shaken daily. The cotton seed and linseed oils developed free acid before they became rancid. On the other hand, olive, rape, castor and arachis (peanut) oils became rancid before their acidity increased. The oils bleached rapidly, castor, rape, arachis and linseed oils became only faintly yellow, cotton seed oil became almost perfectly colorless, while olive oil passed from a yellowish-brown through a bright pale green to a colorless condition. However, it is stated that all olive oils do not bleach with equal ease. As a rule, the specific gravity of these oils increased, while their iodine number decreased. All the exposed oils produced a greater rise in tem-

perature in the Maumene temperature reaction, and a lowering by several degrees of the temperature of turbidity in the Valenta test. The elaidin test was impaired in the case of the olive oil.

Oils which were exposed to the direct sunlight with exclusion of air were bleached but little. The color of oils, stored in the dark, remained unchanged, whether air was admitted to these oils or excluded from them. Therefore, both air and light are required to bleach or change the color of an oil. Rape oil was exposed to light, with exclusion of air, for nine months, then its viscosity was compared with that of another sample of the same oil which had been stored for the same time in the dark in a well-corked bottle. The oil which had been exposed to the light had a slightly greater viscosity.

Fahrion [66] discusses the polymerization of oils. He states that polymerization occurs spontaneously, when oils have stood for some time, and is hastened by warming. The phenomenon is due to the union of the molecules of unsaturated compounds (*e.g.*, glycerides of unsaturated acids) at the double bonds, and therefore, causes a decrease in the iodine number of polymerized oils. Oils which contain a large quantity of unsaturated acids, for instance, linseed, poppy, cotton seed, hemp and nut oils, as well as blubber, are able to polymerize. When air has access to such oils, oxidation may also occur.

Fahrion [67] stored a sample of fish oil for one year in a glass vessel; the cork stopper, which was removed at frequent intervals, was not air-tight. The specific gravity of the oil increased, the iodine number decreased to a great extent, the acidity became greater and the quantity of hydroxy acids showed a slight increase. Fahrion ascribes the changes chiefly to a polymerization at the double bond.

The absorption of oxygen by finely divided oils was studied by Fahrion.[68] He soaked pieces of chamois leather in olive, sesame, rape, cotton seed, poppy, nut, linseed and cod liver oils, and then exposed the leather to the air. With the exception of olive oil, each of the oils absorbed oxygen and reached a maximum gain in weight, then lost in weight. Cod liver oil first attained the maximum gain in weight, then came the drying oils; lastly the other oils. In order to allow for the changing water content of the leather, a control experiment was made upon chamois leather which had absorbed no oil.

The various hydroxy acids, which form when cotton seed, linseed and cod liver oils are oxidized upon the chamois leather, have been identified by Fahrion.[69] Cotton seed oil was absorbed in leather and exposed to the air for several months, during part of which it was in the direct sunlight. The oxidized oil was recovered by means of ether, kept for a half year, then analyzed. The iodine number had decreased enormously; the acidity had increased. The hydroxy acids consisted of di-, tri-, and tetra-oxylinoleic acids. Another sample of cotton seed oil was absorbed in leather and exposed for 14 days at a somewhat elevated temperature; the iodine number of the recovered oil was below the normal value, and mono- and dioxylinoleic acid were present. A thin layer of cotton seed oil was exposed upon a plate to the action of the air for a period of more than a year; the oil then contained trioxylinoleic acid. Apparently tetraoxylinoleic acid marks the limit of oxidation.

Linseed oil was exposed upon leather for 10 days, during part of which the temperature was elevated. The iodine number of the recovered oil had decreased enormously, while the acidity had increased. Di-, tri-, and tetra-oxylinolenic or isolinolenic acids were present. Elementary analysis of a completely dry linseed oil gave a composition corresponding to that of hexa-oxylinolenic acid. However, the dry oil may have been a mixture of oxidation products of linolenic acid, both higher and lower than hexaoxylinolenic acid.

After cod liver oil had been exposed upon leather for five days, it was recovered. The iodine number had decreased greatly while the acidity had increased. Tri- and penta-oxyjecoric acids were present.

Fahrion concludes that, during the oxidation of fats and oils, unsaturated acids with but one double bond usually furnish only volatile and no solid oxidation products. In the case of acids, which contain more than one double bond, one such bond remains intact; the others open up, and an oxygen atom is added at each. Other oxygen atoms enter between carbon and hydrogen with the formation of hydroxyl groups. Polymerization may occur at the intact double bonds; and anhydride-formation may also take place, since hydroxyl groups are present. Different stages of the oxidation may occur at the same time. In the oxidized oils, the hydroxy acids are present as glycerides.

The free hydroxy acids are insoluble in petroleum ether, but dissolve in alcohol.

Spaeth has published analyses dealing with the changes in the same sample of swine fat during a storage of one year [70] and of three years.[58] Swine fat was rendered, filtered, freed from water, analyzed, stored in completely dry glass vessels, which were provided with corks, in a closet in a dry, not too warm, room. After a year had elapsed, the fat was again analyzed. The acidity and refractive index had increased, while the iodine number had decreased.

When the fat had been stored three years, a more elaborate investigation was made. The iodine number of the fat decreased; the acidity, refractive index, acetyl value and Reichert-Meissl number increased, and the melting point became higher. The free fatty acids contained saturated and unsaturated acids in the same ratio in which these acids were present in the original fat. Free glycerol could not be detected. The mean molecular weight of the total fatty acids had decreased. The iodine number of the liquid fatty acids also showed a decrease. After the rancid fat had been washed with hot water, the iodine number of the washed fat was the same as that of the fresh fat. Acetic and butyric acids were present in the rancid fat, which also responded to Becchi's silver solution.

Spaeth states that light and atmospheric oxygen decompose the acids of the oleic series into acids of lower carbon content, hydroxy acids and aldehydes. The decrease in the iodine number of the fat and of the liquid fatty acids, he attributes to oxidation and polymerization. The rise in refractive index is ascribed to polymerization.

Thum [71] separated the free fatty acids from the neutral glycerides of "rancid" olive kernel and palm oils, then liberated the fatty acids of the glycerides. Since both the fatty acids, which had been present in the free state, and those isolated from the glycerides were found to have practically the same iodine number as the oil itself, the conclusion was drawn that saturated and unsaturated acids are liberated in the same ratio in which they occur in the original oil.

Scala,[72] who studied rancid olive oil, decided that rancidity is caused by the oxidizing action of air and light upon the members of the oleic series, especially upon oleic acid. This oxidation of

oleic acid gives rise to formic, acetic, butyric, œnanthylic (n-heptylic), azelaic, sebacic, and dioxystearic acids, and to œnanthol (n-heptyl aldehyde). All these products of oxidation were found in rancid olive oil, and in a sample of pure oleic acid which had been exposed to air and light for nearly a year. The pungent and disgusting odor of rancid oils is ascribed to the presence of œnanthol.

The changes in the constants of certain fats and oils were also studied. Olive oil was kept for 29 months; density, refractive index, saponification number and acidity increased as did the quantity of volatile acids. The iodine number and quantity of fixed (insoluble) acids decreased. The same changes occurred in a lard which had been exposed for 16 months. After butter had been kept for 38 months, the density, saponification number, acidity and quantity of volatile acids had become greater, while the iodine number and quantity of insoluble acids had diminished.

Mjöen [73] drew a current of dry, sterile air, from which carbon dioxide had been removed, through molten sterile butter fat for a period of several weeks. A rancid smell and taste developed, and the fat finally became snow white. The iodine number decreased, while the acidity and saponification and Reichert-Meissl numbers increased; a compound, which reduced ammoniacal silver solution, was present in the oxidized fat.

The chemical process of atmospheric oxidation in direct sunlight differed from that which occurs in ordinary daylight, the sun liberating more acid for the same decrease in iodine number. A butter exposed to sunlight retained its deep yellow color.

After the air current had been drawn through cod liver oil for several weeks, the oil assumed a rancid odor and taste and became more viscous. Light caused the oil to become more or less brown. After a strong current of air had been drawn through olive oil at room temperature for a period of 28 weeks, the oil was practically unaltered. Mjöen states that bacteria and yeasts do not attack and decompose fresh fats.

(*To be continued.*)

MINING & METALLURGICAL SECTION

(Stated meeting held Thursday, November 4, 1909.)

THE MINERAL WEALTH OF THE ISLANDS OF NEWFOUNDLAND AND JAMAICA.

INTRODUCTORY ADDRESS OF THE PRESIDENT OF THE SECTION

A. E. OUTERBRIDGE, Jr.

[The island of Jamiaca is best known as one of the garden spots of the earth and a haven for those seeking to escape the rigors of our winter climate. The extent and accessibility of its mineral wealth are but little known and Professor Outerbridge's account, dealing with the mineral producing localities and their environment, will lend added interest to this region both from the point of view of the tourist and mineralogist.]

It has been my annual custom since my appointment several years ago by the Board of Managers "Professor of Metallurgy of the Franklin Institute," to open the autumn sessions of the Mining and Metallurgical Section with a résumé of Mining and Metallurgical progress during the year. It is my purpose this evening to vary this programme slightly by giving a brief account of some of the mineralogical deposits of two important colonies of Great Britain; namely, the Islands of Newfoundland and Jamaica; one almost bordering on the Arctic Regions, the other in the Tropical Zone.

NEWFOUNDLAND.

At the inaugural meeting of the Mining and Metallurgical Section of this Institute, held April 28, 1897, the paper of the evening, presented by myself, at the request of the President of the Franklin Institute, who occupied the chair on that occasion, was entitled, "The Undeveloped Mineral Wealth of Newfoundland," and was printed in the JOURNAL OF THE FRANKLIN INSTITUTE of September, 1897, embellished with photographic illustrations of the "Iron Mine at Belle Isle, Conception Bay," showing ore chutes and loading pier, and another view showing the surface workings of this mine. Other pictures showed the

"Pyrites Mine at Pilley's Isle" which, even then, employed a fleet of ships. Still another illustration showed the "Copper Mine at Little Bay," which had a total output at that time of about 200,000 tons. Particular attention was paid in this paper to the remarkable iron ore deposits at Wabana, Belle Isle, found near the surface, and the paper stated that "there is little doubt that in future years an extensive export of hematite iron ore will be developed."

How fully that prediction has been realized may be seen by a glance at the following figures, obtained from an interesting paper on "Mining Iron Ore at Wabana, Newfoundland," by B. S. Stephenson, in the *Iron Trade Review* of October 14, 1909.

"During the past ten years there has been shipped from the piers at Wabana over 3,750,000 tons of ore, distributed as follows:

To Germany	1,500,000
To Great Britain	500,000
To United States	1,000,000
To Canada	750,000
Total	3,750,000

Deliveries (to the United States) during the current year will exceed 200,000 tons, sales (bespoken) for 1910 in this country aggregate 250,000 tons and, with the completion of development work already under way on Belle Isle, there is every reason to expect a steady increase in importations from that source."

A few years before the date of my paper the whole of Belle Isle was purchased by the Nova Scotia Steel & Coal Co., Ltd., for a few thousand dollars, and one year later, in 1898, when the Dominion Iron & Steel Company, Ltd., was organized, the lower half of the bed of ore on the land, and a submarine area adjoining the shore, was sold to that concern for a consideration which was reported at that time to have been one million dollars cash. Both companies have since then been operating continually on their respective properties and have now extended their mining operations under the sea.

One year ago the Nova Scotia Company entered on its own submarine areas, after driving a distance of practically 4000 feet. The article in the *Iron Trade Review* says:

"A conservative estimate, made recently by a well known

London engineer, places the amount of ore that may be mined from the land and near submarine areas—after making very liberal allowances for pillar and other losses—at 104,000,000 tons of mineral 'practically proved' and 291,525,000 tons of 'mineral reasonably supposed to exist.' . . . The management of the Nova Scotia Company is fully satisfied that the inner submarine areas alone will yield over 500,000,000 tons of shipping ore, while the total ore underlying the inner and outer submarine areas of this company will exceed 2,500,000,000 tons."

This reads more like a fairy tale than an actual recital of facts and it is within the memory of merchants in St. Johns, New-

FIG. 1.

CLARENDON HILLS, JAMAICA.—The heart of the mining district, showing cultivation of the estates. The copper-wood mine is above the cottage, right hand side.

foundland, who were in the habit of picknicking on the little island and ballasting their sail boats for the return trip across the narrow strip of water, in case of an unexpected gust of wind, with lumps of the ore found lying at the base of the cliffs, when the entire island could have been purchased for a few hundred dollars!

It is evident from what has been here shown, even without referring to other mining operations, that there has been considerable development of the mineral wealth of Newfoundland since the publication in the JOURNAL OF THE FRANKLIN INSTITUTE of my brief paper on its undeveloped mineral wealth.

As I am to be followed this evening by Professor Bradley

Stoughton, who has kindly come here to give us an address on "The Open Hearth Process," I will not take time even to refer to other mineralogical developments in Newfoundland in recent years, but will pass on to a brief account of some observations concerning the undeveloped and partially developed mineral wealth of Jamaica.

FIG. 2.

Highway between the copper mines and shipping port. Old Harbor, 11 miles distant. The military roads in Jamaica are equalled only by the finest park roads in U. S.

JAMAICA.

In March last I visited this beautiful tropical island and saw many evidences of mineralogical deposits of interesting character in the interior and also not far from the coast, but very little had apparently been accomplished in the way of their economic development.

I found that there is a general—if not universal—belief that Jamaica, like many other West India Islands, is of volcanic

origin, but this impression is evidently unwarranted, as will be seen presently.

In the year 1869 there was published in London, by order of the Lords Commissioners of Her Majesty's Treasury, a voluminous and valuable report on the "Geology of Jamaica," by James G. Sawkins, F.G.S.; this is now a rare volume of 339 pages, with numerous tables and maps giving complete accounts of surveys of the entire island; its geological formation, the

FIG. 3.

JAMAICA COPPER MINES.—Open cut showing width of vein between two dark lines. Superintendent and mine boss examining ore.

minerals, rocks, rivers, mountains, etc., and I propose to draw freely from this fund of reliable information for the facts here given. The author says, "The only vestiges of volcanic action occur in the northeast portion of the central range of mountains, below the summit of Blue Mountain Peak (over 7000 feet high) which consists of sand and gravel, represented by quartzite, conglomerate and indurated phosphatic clays."

The island is situated between 17° and 19° north latitude, and 75° and 79° west longitude; it is 135 miles long, 35 to 50 miles broad and comprises 3250 square miles area.

Over 115 rivers and streams are known to find their way to the sea, exclusive of the numerous tributaries from every ravine in the mountainous districts, the abundance of water justifying the name "Island of Eternal Springs" given to Jamaica by the Spanish discoverers. This never failing source of power is practically unlimited but for the most part not utilized.

The following is a condensed list of metallic minerals taken from the geological report referred to:

FIG. 4.

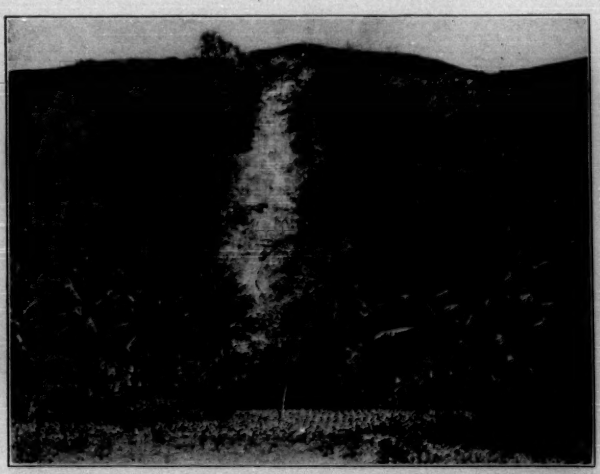
JAMAICA COPPER MINE.—The general manager inspecting a pile of copper ore at the mouth of mine. This ore contains a little gold and several ounces of silver to the ton.

"*Gold*.—This precious metal is found associated with some of the oxidized copper ores of the Clarendon Mines." There is a tradition that the Spaniards obtained gold here and they named an adjacent elevation "The Gold Mine." These workings have long since been abandoned.

"*Copper*.—The ores of this metal are very widely diffused in Jamaica and assume different characteristics according to the geological circumstances and form in which they occur, viz.:

as carbonates in crystals, silicates and even native in granular form. Copper also occurs in veins both in Portland and Clarendon but are most distinctively so in the latter parish. The metal occurs usually in detached deposits among disintegrated rocks on the sides of the mountains either in the form of silicates, carbonates or oxides."

FIG. 5.



COPPER-WOOD MINE, showing opening at top of mountain. Tobacco field and cocoanut grove in the valley. Jamaica.

In the geological report of the parishes of St. Thomas and Portland no less than twelve pages are devoted to descriptions of the various copper deposits. In the chapter entitled "Economic Geology of the Parish of Clarendon" the following account appears: "The Clarendon Mines afford a nearer approach to true lodes of mineral veins than any of the other metalliferous deposits of Jamaica. Without being contained in mechanical fissures, still, the deposits are bound by definite walls and characterized by distinct gangue and ribs of ore. . . . During the year 1858 some specimens of blue and green carbonates of

copper were given to the Hon. Lewis Mackinnon as specimens of copper ore from the Clarendon mines and on leaving Halse Hall these specimens were thrown away. While I was surveying this (Clarendon) parish I occupied Halse Hall, and the Assistant Geologist, Mr. C. B. Brown, while on a visit, found a small piece of the discarded rock and on close examination discovered that what had been considered by the miners copper pyrites was truly

FIG. 6.

PAY DAY AT THE MINES.—The negroes receive about 36 cents per diem. There are 150 men working, night and day shifts. Jamaica copper mines.

gold. Large pieces were afterwards found containing sufficient gold to be considered gold ore, the value was established at from £60 to £70 per ton, besides the copper which would yield from 12 to 15 per cent."

It is of record in the archives of the colony that in the year 1857, 207 tons 15 cwt. of copper ores were shipped from the Clarendon mine and Mr. Sawkins states that had these mines been worked intelligently and due attention given to the under-

taking, the mining would have then proved a success instead of a failure. It is the same region that is now being developed by Boston capitalists and is in the centre of an agricultural district under high cultivation.

In Appendix III of Mr. Sawkins's report there are quite a

FIG. 7.

Free hand sketch of narrow trail to top of Blue Mountain Peak, Jamaica, (7443 feet elevation), showing one of many sharp turns on dizzy heights with conical peaks below. The trip required two days of hard riding on sure footed mountain ponies.

number of analyses of evidently selected specimens of copper ores from different localities, among which are the following:

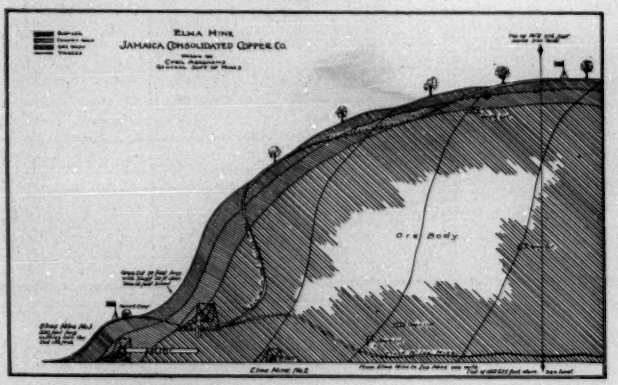
	COPPER
Copper Pyrites	35.93 per cent.
Purple or Peacock ore	62.50 per cent.
Mixed purple and gray ore	70.99 per cent.
Gray copper ore	79.80 per cent.
Native copper	98.97 per cent.

Among other minerals described in the report as being found in Jamaica are cobalt, lead (galena), zinc (sulphide), manganese (pyrolucite), iron, arsenic, and antimony. No mining operations have been established so far as I have been informed, except for

copper in the Clarendon hills where extensive developments are now in progress.

In 1906 several Boston men obtained a lease of 2276¾ acres of land in these hills, with option of purchase within a few years, and since then have been vigorously prospecting and tunnelling into the hills. At the time of my visit to Jamaica (March, 1909) there were about seventy-five laborers at work blasting out ore and sixty-two openings had been made, from more than forty of which it was said that different kinds of ore had been taken showing profitable copper values and carrying more or less value in silver and gold.

FIG. 8.



Sketch of Elma Mine—one of ten mines now opened on the property.

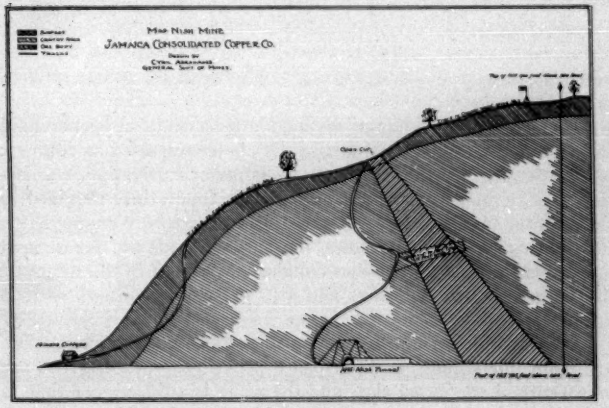
I secured a collection of samples of these copper ores from several of the mines and they have all been classified at the Academy of Natural Sciences. Duplicate samples, duly labelled, are on the table for inspection at the close of the meeting.

Several of the specimens are very rich ores. I have also obtained copies of forty-seven analyses made by six different firms in New York, Boston, Denver, Jamaica, and London, of samples taken from the run of the mines that have been analyzed for the purpose of obtaining definite information regarding the average value of the deposits, these show, of course, wide variations, ranging from a minimum of 4.06 per cent., to maximum

69.32 per cent. copper, also showing gold, ranging from a mere trace up to 1.72 ounces per ton, and from a trace of silver up to 56.44 ounces per ton of ore.

At the time of my visit in March last, a little over 1200 feet of tunnels had been made, since that time the force of laborers has been doubled and development work has progressed rapidly, as

FIG. 9.



Sketch of MacNish Mine, recently opened.

will be seen from the annual report of the General Manager of the Jamaica Consolidated Copper Co., dated October 13, 1909, just issued.

"I am pleased to report most satisfactory progress at the Mines during the past twelve months. Our development now stands as follows, in tunnels, cross-cuts, upraises and drifts:

	FEET
Sylvia Mine	819
Victoria Mine	205
Cheltra Tunnel	403
Iva Mine	341
Elma Mine	384
Clarissa Mine	156
Copper Wood Mine	557
Cyril Tunnel	47
Congo Hill Mine	1271
MacNish Tunnel	173

making a total of 4356 feet, with more than 2000 feet of open cuts and prospect shafts not included in the above.

"The Mines have recently been examined by one of the ablest mining men in America. He reports as follows:

Ore in sight ready for extraction,—	TONS
Congo Hill	1,332,000
Victoria Mine	66,500
Sylvia Mine	150,000
Iva Mine	19,800
Elma Mine	16,500

with all breasts and drifts in ore. This makes a grand total of 1,585,800 tons."

Some of the peculiar or distinctive features of copper mining in the Clarendon hills in Jamaica may be enumerated as follows:

1. A great portion of the estate is under cultivation, furnishing considerable revenue, besides all the food stuffs required to feed all the hands now, or that may be in the future employed on the plantation or in the mines, the balance is mainly forest land containing all the timber that could ever be used in the mines or in the erection of buildings and also precious woods of various kinds.

2. Native labor is employed with success at an average wage of about one shilling and six pence per diem. The negroes are apparently a contented class and the wage is a little more than is paid agricultural labor on the neighboring plantations.

3. The distance from the shipping point "Old Harbor," or from the railway station, "May Pen," is a little more than 10 miles over highways so perfect as to be compared only to park roads.

4. The climate is such that work can be carried on day and night throughout the year.

5. Abundant water is available for power and for electrolytic refining operations at low cost.

The following classification of the samples of copper ores from seven different mines on the property of the Jamaica Consolidated Copper Company has been kindly furnished by the Curator of the Academy of Natural Sciences. Duplicate specimens have been presented to the Academy and to the Franklin Institute.

No. 1.—Bornite with azurite and malachite; from Iva Mine.
No. 2.—Chalcocite with malachite and chrysocolla; from Victoria Mine; very rich ore.
No. 3.—Chrysocolla and chalcocite; from Sylvia Mine; rich ore.
No. 4.—Bornite and malachite with chalcocite and chrysocolla; from Clarissa Mine.
No. 5.—Metallic mineral, dorneykite or mohawkite; red portion, cuprite (copper oxide); from Sylvia Mine.
No. 6.—Chalcocite and chrysocolla; from Elma Mine.
No. 7.—Red part, cuprite; blue green portion, chrysocolla; bright green, garnerite; tarnished patches, Mohawkite. (The garnerite is nickel and magnesium silicate.)

In conclusion I think it proper to repeat what I said in my address given in 1897; namely, that I am merely an observer from an outside standpoint and chronicler of these exploitations, and am not connected in any capacity with any of the mining companies. I am not, therefore, prepared to express an authoritative opinion as to the future commercial value of any of the operations described, but I have verified, as far as possible, statements furnished to me before presenting them in this paper. Should the development of the mineral wealth of Jamaica in the next decade compare favorably with that of Newfoundland since my address in 1897, I hope I may have the pleasure ten years hence of calling attention to it, as I have just now done with respect to the remarkable developments of mineral wealth of Newfoundland in the past twelve years.

NOTE.—At the conclusion of this address a number of lantern slides made from kodak pictures taken by the author and his party were exhibited showing the general character of the country, together with views up the trail leading to the top of Blue Mountain Peak. This is a difficult and rather dangerous trip, occupying two days of very strenuous riding; skirting numerous precipices and affording magnificent views. Since the earthquake of 1907 few, if any, visitors to the island have made this ascent, and the path is somewhat overgrown and partly obscured in many places. It is necessary to employ guides also to carry warm clothing, and to arrange in advance for accommodation over one night, or possibly longer, in case of bad weather, at one of the coffee plantations on the side of the mountain. It is not always possible to obtain such shelter.

MINING & METALLURGICAL SECTION

(*Stated meeting held Thursday, November 4, 1909.*)

RECENT PROGRESS IN OPEN HEARTH STEEL PRACTICE.

BY

BRADLEY STOUGHTON.

[The basic open hearth furnace, the world's greatest steel producer at present, suffers under the disadvantage of greater cost of operation than the Bessemer converter, due chiefly to costs for fuel, labor and repairs. Constant experiments are being made at steel works to reduce these expenses, and the important modifications in design and practice that have resulted are discussed in this paper, together with some reference to the attempts to make low-carbon steel of high quality in the open hearth furnace, and brief notes on the Duplex process.]

THE operation of the open hearth furnace (see Fig. 1) is too well known to you to require any description, and I wish to confine my attention to-night to recent innovations in furnace design or operation which have fought their way to notice and maintained a position after the test of practical experience.

Fuel.—Producer gas is still the most important fuel used, because it gives more heat units for a dollar than any other manufactured fuel, and it is even increasing over natural gas owing to the diminishing supply of that product in the Pittsburgh district. With coal at two dollars per ton and natural gas at ten cents per thousand cu. ft. the cost as open hearth fuel is approximately the same for the natural and the manufactured product, provided the gas producer plant is large enough to be operated at economical figures. One advantage of natural gas in this comparison is its high calorific power per cu. ft. which enables it to be used without passing through any regenerative chamber.

In England and Europe, where they are more progressive in some departments of the steel industry, or, better say, where they are progressive at an earlier period of development of some improvements in the industry, the use of spare gas from by-product coke ovens is established in open hearth practice, but some American manufacturers believe that this particular form of progressiveness is still a little premature. Coke oven gas is

so rich as not to require regeneration, and is obtained as a by-product at a low cost.

Fuel oil costs initially more than the equivalent amount of coal, but can be transported much cheaper. Therefore, its use depends chiefly upon location. About 40 gallons of oil will do the work of producer gas from 500 lbs. of coal, or else of 6000 cu. ft. of natural gas, and will cost nearly twice as much in Pittsburgh at present prices. Oil requires no regeneration, gives a more oxidising flame which can be controlled easier than gas, is not so damaging to the roof of a furnace, but more so to the walls. Heats can be brought out slightly quicker with oil fuel, but the steel is more liable to be unsound unless great care is employed.

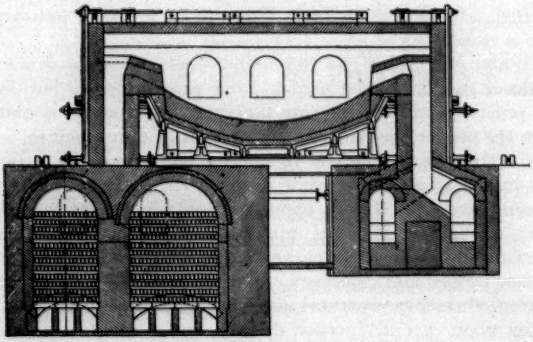
Water gas, Mond gas and similar fuels of higher grade than producer gas, have been used to some extent abroad, but American practice seems to indicate that greater economy is obtained from the poorer fuel and simpler process of manufacture.

Furnace Repairs.—The rebuilding and repairs of a furnace cost in a year about the same as the total fuel used, depending to some extent, of course, upon how location affects the relative prices of coal, refractory materials and labor. Also these renewals per year are not very far from the initial cost of furnace proper and chambers. Their magnitude, as can readily be seen, therefore warrants the expenditure of both time and money upon careful furnace design, and every year sees some improvements in this respect. Within a few years the number of heats that a basic furnace would make before it became necessary to cool it down for repairs has increased by from 50 to 100 per cent., and at the same time repairs between heats has decreased slightly.

The necessity for repairs arises from four causes: (1) the corrosive action of metal and slag upon hearth and brick-work; (2) the softening and melting of the brick-work by heat; (3) cracking of the brick-work by expansion and contraction with the heat; and (4) choking of the spaces between bricks in the regenerative chambers by soot, or by dirt and slag thrown over from the hearth. The relative importance among these of the corrosive action of the slag is illustrated by the circumstance that the furnace with a basic slag requires to be cooled down for repairs nearly three times as often as that with an acid slag.

This is due in part directly to the corrosive action of the basic slag and in part to the volume of this slag which makes a heavier blanket between flame and metal and thus necessitates more heat in the furnace, from which the brick-work suffers. From this it must not be inferred that the cost of rebuilding and repairs in the basic furnace is quite three times, or even twice, that in the acid furnace, because, although the life of the basic furnace is shorter, the extent of rebuilding in the acid furnace is much greater when it does become necessary.

FIG. 1.

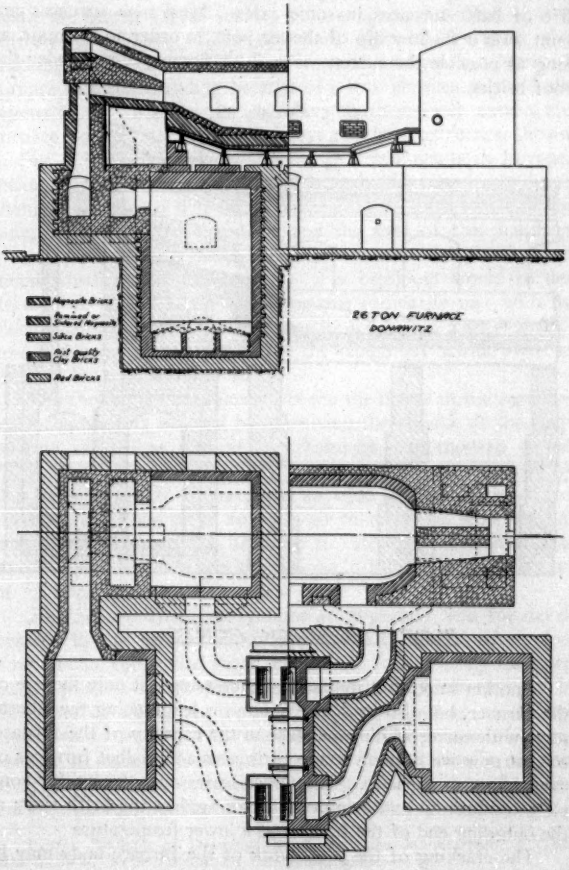


Siemens open-hearth furnace.

The destruction of brick-work by corrosion and melting has been lessened by several recent devices, following the early ones of supporting the roof substantially on I-beams and air-cooling the hearth by leaving it open as much as possible outside. In the latter respect the Europeans have gone further than we have as regards the brick-work near the up-takes, and the up-takes themselves and ports, as shown by a comparison of Figs. 2 and 3, but American practice now places an air-cooled cast block at the corner of the hearth-pan next the ends, which is effective in protecting this location. A layer of chrome ore on the floor of the ports and, in basic practice especially, other exposed places where slag is liable to lodge and corrode is a frequent safeguard. Ports are sometimes designed (see Fig. 3) so that the relatively

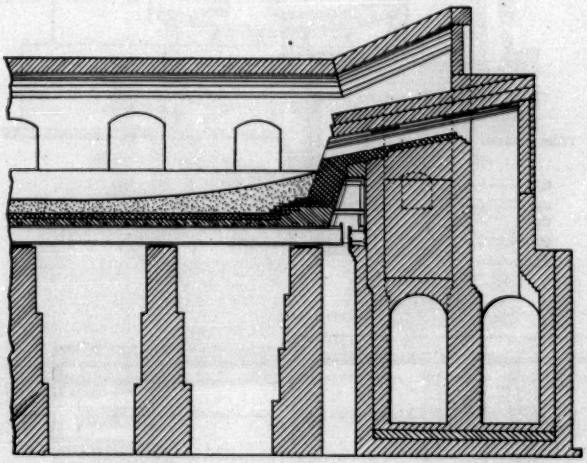
fragile wall between them is exposed to both of the outgoing currents of gases for only a part of its length, and thus will

FIG. 2.



retain almost indefinitely enough length to give direction to the incoming gas and air when their current is from that side. Still more recently, the port and bulkhead have been water-cooled by patented devices, and this practice is said to have doubled the life of basic furnaces in some cases. Most new furnaces are built with a shallow dip of the air port, in order to maintain as long as possible the current of cool air flowing along under the roof bricks.

FIG. 3.



BLAIR FIREBRICK PORTS

Another important innovation, increasing not only the life of the furnace, but also its fuel economy, is lengthening the laboratory, with corresponding increase in the capacity of the furnace, so that now we hear of 100-ton furnaces and Talbot furnaces of even 225 tons capacity. This gives better opportunity for complete combustion and brings the gases against the brick-work at the outgoing end of the furnace at a lower temperature.

The cracking of the brick-work of the furnace body may be

materially lessened by careful attention to the adjustment of tension on the structural tie-roads by the turnbuckles now always provided for that purpose, and is also met with less frequently since the practice of building the regenerative chambers underneath the hearth (these serving as a support) has been almost entirely abandoned. This departure likewise decreases the cracking of the chambers themselves, and we hear less often of furnace campaigns ending because of a leak between gas and air regenerators. Placing the chambers farther away makes the furnace work a little more sluggishly, and the construction shown in Fig. 2,* which has been adopted at a few plants in Europe, makes it possible to avoid this objection. If this design were changed slightly so that the hearth would be supported entirely independently of the chambers, and the roof of the chambers given a pitch towards the back of the furnace so that the flood of metal which would occur in case of a break-out would be deflected, we should avoid the objections ordinarily met with by placing regenerators under the hearth, and would gain in quickness and compactness and escape the danger of a leak between gas and air regenerators.

The choking of the spaces between the bricks in the regenerative chambers is lessened by increasing the volume of the slag-pockets as well as that of the chambers. In modern 50-ton furnaces it is very common for the volume actually occupied by the checkerwork to be as much as 1900 cu. ft. for each gas chamber, and 2600 cu. ft. for each air chamber, while its vertical height will be as much as 18 to 20 ft. Perhaps more consideration has been paid to these dimensions in European practice than in America, except within a few years.

Soft Steel.—There yet remains an important field for development in the production of low-carbon steel for wire, pipe, rivets, bolts, cold-rolled stock, blacksmith iron, etc., by the open hearth process. Each year there are more furnaces engaged in this class of work, but the steel, so far at least as all I have examined, is charged with oxide of iron. The process of manufacture is usually to melt a charge consisting chiefly of steel scrap, the small proportion of metalloids in which is oxidised out during melting: or else to maintain an oxidising slag up to the

* From "Album of Drawings Relating to the Manufacture of Open Hearth Steel," by A. Pavloff, St. Petersburg, 1908.

moment of tapping. The wild steel resulting from both of these methods is passable only when the carbon, phosphorus and sulphur are very low, and when the consequent blowholes are effectively welded up during rolling. The success obtained by manufacturers in this class of product is an evidence of the constant care and skill exercised, but there is still much reluctance to entering very far into this class of manufacture with the open hearth furnace.

Duplex Process.—Probably the one development in steel manufacture that is attracting the most attention at the present time in America is the increasing use of the Duplex process in large steel plants. The slowness of the open hearth operation and the rising price of suitable melting scrap combine to enhance the value of a method which will improve the former and avoid the latter. There is still much variation in the details of this combination of the Bessemer and open hearth processes so as to obtain the quickest and best results, even in the same plant at different times, and it will probably be some years before a standard of practice will become established, while the mechanical details of handling the molten metal and transporting it from blast furnace to Bessemer converter and open hearth, and from converter to open hearth, either with or without the intervention of one or two 500-ton or 750-ton reservoirs, are being improved yearly. To-day we find some plants using together in the open hearth furnaces both liquid pig iron and liquid blown metal from the converters, while others will use blown metal only. Some plants are using alternately each type of combination, until experience proves which is better. Where blown metal is used alone it is customary not to reduce the carbon in the average charge for any open hearth furnace below 1 per cent., and this carbon is oxidised in the hearth, together with the phosphorus and part of the sulphur. Even when the blown metal is mixed with liquid pig iron it is not always blown dead. At present the Duplex process is not only practiced with the ordinary open hearth operation, but in Talbot furnaces as well, and promises to give material success.

NAVIGATING THE AIR.

BY

AUGUSTUS POST,

Secretary, Aero Club of America.

RÉSUMÉ OF THE POPULAR SCIENCE LECTURE DELIVERED AT WITHERSPOON HALL, FRIDAY EVENING, OCTOBER 15, 1909.

The lecture was prefaced with a brief introductory address by President Clark. The speaker pointed out that aerial navigation had been developed by the evolution of the three distinct classes of apparatus—balloons, dirigibles, and aeroplanes; and presented the subject in the following order:

Balloons.—Successful navigation of the air may be fairly said to have its first impetus with the experiments of the Montgolfier brothers. Their crude experiments on the ascentional power of "smoke" and "steam" finally led to the use of heated air as a means of securing buoyancy. Shortly after this, through the investigations of the brilliant physicist Charles, hydrogen was found more suitable and balloons, even at that early date, barring advances in construction and other technical details, differed little from those used at present.

At that time there existed many technical difficulties in the manufacture of the gas which have been overcome in modern generators. Among the improvements in modern gas plants may be mentioned a steel cylinder for convenient transportation of gas at high pressure to the field of operation.

The principal requirements in balloon construction are—envelope must be gas tight and water-proof, the netting must properly fit the envelope, and the car must be suspended in a certain position fixed by the shape and dimensions of the gas bag.

Among the methods of control may be named the gas valve at the top of the envelope for controlling the escape of gas from the car, the ballast, the drag rope and the anchor, all of these seemingly crude methods of control permit navigating a balloon, by taking advantage of air currents at different elevations, with a degree of precision surprising to the uninitiated.

An equipment of instruments to aid navigation is provided, such as the compass, the recording barometer, and the anemometer. Among these there is one peculiar to ballooning known as the "stotoscope" which utilizes the change of barometric pressure, due to a change of elevation, to indicate by the motion of a delicate diaphragm whether the movement is one of descent or ascent.

Dirigibles.—Although attempts to construct balloons propelled by mechanical means were made over fifty years ago, it is only within comparatively recent years—since the advent of the liquid-fuel internal-combusion motor—that such efforts have met with practical success. The envelopes are invariably elongated in a horizontal direction, to present as small a cross-sectional area as possible in the direction of motion. The best known and most successful of these are those built by Lebaudy Parseval, de la Vaulx, and Zeppelin. With the exception of the last these all have a flexible envelope like a balloon. Zeppelin differs from the others in having a rigid envelope and is a most interesting achievement in extremely light construction.

Dirigibles are without exception, exceedingly susceptible to injury on account of their large volume and extreme lightness when anchored, by any sudden gust, or, other inclemencies of weather, and in order to protect them they are stored, when not in use, in air-ship houses. These are large

frame structures capable of swinging about a pivot in order that the opening may be made to face various points of the compass according to the direction of the wind.

Aeroplanes.—Ascentional power in aeroplane apparatus is secured entirely by the application of mechanical power; the vehicle itself, though light, being quite devoid of buoyancy in air. The principle utilized is that when a plane, or a slightly curved surface, is driven through the air in a direction having small inclination to the direction of motion, the component of air pressure perpendicular to its direction of motion is far greater than that required to drive it. Much theoretical investigation and many experiments have been made to determine the more effective size, shape, and arrangement of an aeroplane to obtain a maximum lift. Notable among the modern experimenters in this field may be particularly named Maxim, Langley, and Lilienthal. The methods of the latter are said to have been followed by the Wrights, the early trials of which culminated in their undisputed preëminence. Their successful efforts have been quickly followed by equally remarkable flights by Farnum, Curtis, Bleriot and others. Aeroplane production is now an established industry in Europe and equipment and motive power peculiar to this type of vehicle may now be purchased, which, in refinement and adaptability, is a delight to the mechanical instinct.

[Mr. Post elaborated extensively upon all the topics mentioned in this résumé and illustrated the subject by a large number of lantern slides and moving pictures depicting in a most striking manner the appearance of dirigibles and aeroplanes in operation.]

TITANIUM.

USES OF A RARE METAL.

The use for titanium which gives promise of greatest expansion is in steel and cast iron. In steel it is ordinarily added in the form of ferrotitanium (containing preferably from 10 to 20 per cent. of titanium) in quantity sufficient to form about 0.1 per cent. of the steel. Rails treated with titanium and laid in places on railroads where the wear was especially hard are said to have shown much less wear than untreated rails. Gray cast iron also is said to have shown beneficial effects from such treatment. Cuprotitanium is also manufactured for use in bronze and other castings containing copper. The titanium acts as a deoxidizer, much like phosphor-tin, and makes very tough castings. Another use of titanium which promises to assume considerable proportions is the manufacture of electrodes for arc lights.

Titanium ore in the form of rutile, which is titanium dioxide, has been mined in Nelson County, Virginia, where a plant has been erected to extract the metal from the ore. Titaniferous iron ores are mined in New York, Minnesota, and Wyoming.

A report on the production of titanium and other rare metals in 1908, prepared by Frank L. Hess, has just been issued by the United States Geological Survey.

FRANKLIN INSTITUTE

(*Proceedings of the stated meeting held Wednesday, November 17, 1909.*)

HALL OF THE FRANKLIN INSTITUTE,
PHILADELPHIA, November 17, 1909.

President WALTON CLARK *in the Chair.*

Addition to membership since last report, 5.

The following members were elected to the Committee on Science and the Arts to fill unexpired vacancies: To serve one year and two months—Carl L. E. Schwartz; to serve two years and two months—Dr. Henry Leffmann, and Dr. Robert H. Bradbury.

The paper of the evening was read by Dr. Leo H. Baekeland of Yonkers, New York, on "Bakelite and its Applications."

Bakelite is a liquid compound, having as a base, phenol and formaldehyde, which on solidification resembles amber in appearance and when used in combination with wood, paper, asbestos, graphite, and other substances, produces a solid, non-resistent, non-inflammable insulating material which will have many applications in the various manufactures. The subject was illustrated by experiments and specimens of the product.

At the close of the meeting it was moved and seconded that the subject be referred to the Committee on Science and the Arts for investigation and report. After a brief discussion the thanks of the meeting were extended to the speaker for his interesting address. Adjourned.

JAMES CHRISTY,
Secretary.

COMMITTEE ON SCIENCE AND THE ARTS.

(*Abstract from proceedings of the stated meeting held Wednesday, November 3, 1909.*)

HALL OF THE FRANKLIN INSTITUTE,
PHILADELPHIA, November 3, 1909.

MR. J. Y. MCCONNELL *in the Chair.*

The following reports were presented for final action:

(No. 2432.) Color Photography.—Auguste and Louis Lumiere. Adopted, Elliott Cresson Award.)

(No. 2457.) Telelectric Piano Player.—John F. Kelly, Timothy B. Powers, and Major R. Jewell. (Adopted, John Scott Award.)

R.

BOOK NOTICES

BIBLIOGRAPHY OF THE COTTON MANUFACTURE. By C. J. H. Woodbury, A.M., Sc.D. Consulting Engineer. Secretary of the National Association of Cotton Manufacturers, Boston, Mass. 213 pages, frontispiece, 8vo. Waltham, Mass, E. L. Barry, 1909.

The volume contains over 5000 references to the literature of cotton manufacture and related subjects.

Works in many languages are included—we note titles in English, German, French, Spanish, Italian and Norwegian.

Ten pages are devoted to an extensive list of periodicals, directories and dictionaries.

It is surprising to learn that so much has been written on the subject—there are 820 entries under the heading Cotton Manufacture, and 874 titles in the division Finishing.

Librarians and persons interested in textile matters will find the book useful for reference. R.

MESURES ÉLECTRIQUES—ÉTALONS ET INSTRUMENTS, ESSAIS MÉCANIQUES, PHOTOMÉCANIQUES, MAGNÉTIQUES ET ELECTRIQUES—APPLICATIONS AUX LIGNES, GÉNÉRATEURS, MOTEURS ET TRANSFORMATEURS. Leçons données a l'Institut Électrotechnique Montefiore par Eric Gerard, Directeur de cet Institut, Ingénieur principal de télégraphes, Professeur a l'Université de Liége. Third edition, ix + 702 pages, 9 x 6 inches with 304 figures. Price, paper covers, 12 francs. Gauthier-Villars, Paris, 1908.

The extensive field covered by this treatise as indicated in the title, "Methods of Procedure Evolved from Theoretical and Practical Considerations," and the description in detail of apparatus, make it not only efficient reference work for the experimenter but also a valuable text for the systematic study of the subject.

The work, now in its third edition, has been thoroughly revised to keep pace with the present-day rapid progress and refinement of electrical science. Professor Gerard's "Leçons sur l'Électricité" is widely known among English-speaking electrical engineers to which this more recent work by its eminent author constitutes a worthy sequel. L. E. P.

CANADA, DEPARTMENT OF MINES, MINES BRANCH. Report on the Chrome Iron Ore Deposits in the Eastern Townships, Province of Quebec. By Franz Cirkel, M.E. 141 pages, illustrations, plates, 8vo. Ottawa, Government Printing Office, 1909.

In the last few years, owing to the rapid development of the iron and steel industry in Canada, more than ordinary interest has been manifested by the mining and metallurgical public in respect to the Canadian deposits of raw materials and ores, which enter into the manufacture of iron and steel products. To meet the demand for information on these subjects, the Mines Branch has undertaken to issue a series of reports on the Canadian deposits of the substances which form the basis of the iron and steel industry. Several reports on iron ores and one on tungsten ores have already been issued and the present report is another of this series.

The scope of the report on the chrome iron ore deposits is best judged by quoting the Table of Contents, which is as follows:

"Introduction; Chapter I, Historical; Chapter II, The Chrome Iron

Ore Deposits of Canada; Chapter III, Mining of Chrome Iron Ore; Chapter IV, Dressing for the Market; Chapter V, Market Prices, and Status of the Canadian Industry; Chapter VI, Chrome Iron Ore Mines—Prospects in Canada; Chapter VII, Chrome Iron Ores in Foreign Countries; Chapter VIII, Origin; Chapter IX, Composition of Chrome Iron Ores; Chapter X, Statistics and Chronology; Chapter XI, Determination of the Value of Chromium; Chapter XII, Uses of Chromium; Chapter XIII, Technology of Chromium and its Compounds; Appendix II, Experiments with Chromite at McGill University; Bibliography."

The report is well illustrated by plates in half-tone, and diagrams, maps and drawings. The author of the report, Mr. Fritz Cirkel, Mining Engineer, was entrusted with the work on account of his long connection, as consulting engineer, with the chromite and the asbestos industries. The deposits of these two substances in the province of Quebec occur in the same region, and the origin of both is closely connected with the serpentine rocks of the Eastern Townships. R.

TABLES AND OTHER DATA FOR ENGINEERS AND BUSINESS MEN. Compiled by Chas. E. Ferris, B.S., Professor of Mechanical Engineering, University of Tennessee. Published by the University Press, Knoxville, Tenn. Thirteenth edition, 165 pages, 3 x 6 inches, leather. Price fifty cents.

The avowed object in preparing this manual is to secure a medium whereby the strongest possible arguments in favor of technical education as a means of developing our resources may be brought before the minds of men who control the affairs of the South. Pursuant of this object, the work is prefaced by an account of the organization, the courses, and opportunities at the university. The subject matter is similar to that ordinarily found in the usual standard reference books devoted to mechanical engineering, though necessarily of much less scope, due to its diminutive volume. A book of this kind within convenient reach should receive frequent consultation and prove an efficient vehicle in promoting and extending the work of the university. L.E.P.

ELEMENTARY ELECTRICAL CALCULATIONS: A manual of simple engineering mathematics, covering the whole field of direct-current calculations, the basis of alternating-current mathematics, networks and typical cases of circuits, with appendices on special subjects. By T. O'Conner Sloane, A.M., E.M., Ph.D. x + 304 pages, 5 x 8 inches, cloth. D. Van Nostrand Company, New York. Price, $2.00 net.

The scope of this book is described in the title. This purpose stated by the author is to give in simple form what may be termed a foundation for the study of electrical calculations. It will probably find its greatest value, first, among beginners who have a reasonable working knowledge of elementary mathematics and, again, among a large class of engineers, who, exclusively devoted to practice along other lines of industry, will be glad to have at hand a book in which the process of solution of an electrical problem may be quickly found. The book is well arranged for systematic study and contains a generous number of classified problems at the end of each chapter. It should appeal to all who require a thoroughly practical course in elementary electric calculations. L.E.P.

ERRATUM

November issue, page 397. La Théorie des Courants Alternatifs; for volume II read volume I.

POPULAR SCIENCE LECTURES.

The JOURNAL desires to call the attention of its readers to the Course of Popular Lectures being given this winter at Witherspoon Hall, Walnut and Juniper Streets, Philadelphia, under the auspices of the Franklin Institute.

The first lecture on October 15, was by Mr. Augustus Post, Secretary of the Aero Club of America. His subject was entitled "Navigating the Air." An audience of upwards of 1000 people listened to this interesting lecture which was illustrated with lantern slides showing the various types of Air-ships in motion, as well as descriptive slides of the Aviation Meeting held last summer at Rheims, together with other lantern projections of the most important parts of flying machines.

On Friday, November 12, the second lecture of the Course was given by Mr. Paul F. Bauder, of Cleveland, Ohio. Mr. Bauder is identified with the National Electric Lamp Association and well qualified to speak on the subject matter of his lecture, "The Quality of Light." The lecture was profusely illustrated by colored lantern slides and other pictures descriptive of his subject.

The next lecture will be on Friday evening, December 10, and the subject chosen is entitled "Perils of Peace, or a Safer America." The lecturer is Dr. W. H. Tolman, Director of the Museum of Safety and Sanitation, and one of the Delegates to the International Medical Congress held last summer at Buda-Pesth, where he presented a paper on the scope and object of his Museum which received the strong endorsement of the Congress. In this country he has the support and encouragement of such well-known men as Elbert H. Gary, Richard Watson Gilder, Philip T. Dodge, Thomas Darlington, and many other equally prominent and well-known gentlemen.

On the evening of Friday, January 28, 1910, Mr. Logan W. Page, Director of the Office of Public Roads, Washington, D. C., will give an interesting address on "*Road Administration and Maintenance.*" The speaker, who is thoroughly conversant with the details of highway construction and maintenance, will describe the work which is being carried on at home and abroad in this timely branch of engineering.

Dr. Robert H. Bradbury of the Southern Manual Training High School of Philadelphia will speak on Friday, February 11, 1910, on the "*Recent Methods for the Production of Light.*" During the course of the lecture the recent illuminants—the Welsbach light, tantalum, tungsten, and Nernst lamps, the Macfarlan Moore vacuum tube, the Cooper-Hewitt lamp and the flaming arc—will be shown in operation.

The closing lecture of the season will be held on Monday, March 14, 1910. On this occasion Messrs. W. N. Jennings and Charles K. Stokes will describe their most recent journey and will show a series of new photographs. Mr. Jennings has taken part in the Popular Science Courses for many years and is known to all the members of the Institute.

It is the earnest desire of the Committee on Instruction that the members and friends of the Franklin Institute will use their endeavors to give these lectures a large attendance. The subjects are all attracting much attention just at present throughout the entire country and the qualifications and standing of the speakers should insure well-filled houses. Members of the Franklin Institute should avail themselves of the privilege of their membership-cards and attend these lectures. They will all be illustrated with lantern slides, and Witherspoon Hall, in which they will be given, is most attractive and comfortable. *Tickets can be had by applying at the Institute.*

THE EDITORS.

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INDEX TO VOL. CLXVIII.

Aerial navigation (Post), 477
Automobiles, gasoline, electric (Entz), 57

BALCH, EDWIN S.: Develop the Submarine! 108
Balloon, dirigible, with gyroscope control (Thayer), 19
Baltimore, Power-house economics in (Foster), 315
BIRKINBINE, JOHN: The Mixteca country in the State of Oaxaca, Mexico, 200
BOOK NOTICES:
Barr, J. R.: Principles of direct current electrical engineering, 81
Boyer, J.: La synthèse des pierres précieuses, 313
Brunswick & Aliamet: Construction des induits a courant continu, 81
Canada, Department of Mines: Investigation of the peat bogs and peat industry of Canada, 312
Canada, Department of Mines: Report on the chrome iron ore deposits, 480
Canada, Department of Mines Report on iron ore deposits of Nova Scotia, 312
Chaplet A.: Les succédanès de la soie, 82
Chatelain, E.: Soudure autogène et aluminothermie, 398
Ferris, C. E.: Tables and other data for engineers, 481
Gerard, E.: Mesures electriques, 480
Gerhard, Wm. P.: Sanitation and sanitary engineering, 153
Granderye, L. M.: Determination des roches, 153
Haenig, A.: Die steinkohle, 398
Heess, J. K.: Practical methods for the iron and steel works chemist, 313
Houghton, C. E.: The elements of mechanics of materials, 398
Pontio, M.: Analyse du caoutchouc et de la gutta-percha, 397
Russell, A.: La théorie des courants alternatifs, 397
Sidersky, D.: La réfractométrie et ses applications pratiques, 397
Sloane, T. O'C.: Elementary electrical calculations, 481
Soliman, G.: Étirage, tréfilage, dressage des produits métallurgiques, 81
Tower, O. F.: A course in qualitative chemical analysis of inorganic substances, 153
Woodbury, C. J. H.: Bibliography of cotton manufacture, 480
BRADBURY, ROBERT H.: Recent progress in the chemistry of the proteins, 85
Building operations in 1908, 130

CARTER, OSCAR C. S.: The interior of the earth in the light of the new seismology (correspondence), 303
Coal, Effects of oxygen on, 335
Coal, run-of-mine and briquetted, on locomotives, 218
Compass, card, reduced diameter, 300
CORRESPONDENCE:
Earthquakes in the light of the new seismology (Hixon), 227
The interior of the earth in the light of the new seismology (Carter), 303

Earth, interior of the, in the light of the new seismology (Carter), 303
Earthquakes in the light of the new seismology (Hixon), 227
Electric furnaces in Germany, 104
ELY, OWEN: Newton's law and the cause of gravitation, 121
ENTZ, JUSTUS B.: Gasoline electric automobiles, 57
Eucalyptus poles, Government studies method of seasoning, 226

Fats and oils, A critical study of the natural changes occurring in (Hepburn), 365, 421

Forestry, Some problems in (Seely), 1
FOSTER, HORATIO A.: Power-house economics in Baltimore, 315
FRANKLIN INSTITUTE:
Committee on science and the arts: Abstract of proceedings of the stated meeting, June 2, 1909, 80; September 1, 1909, 311; October 6, 1909, 396; November 3, 1909, 479
Committee on science and the arts: Gayley dry air blast, 67
Committee on science and the arts: Vernaz milling files, 219
Popular science lectures, 482
Proceedings of the stated meeting, June 16, 1909, 81; September 15, 1909, 311; October 20, 1909, 396; November 17, 1909, 479
FRAZER, PERSIFOR: Obituary notice, 75
FULWEILER, W. H.: The development of modern road surfaces, 155, 260

Gayley dry air blast (Franklin Institute report), 67
Gravitation, Newton's law and the cause of (Ely), 121

Hardwoods, eastern, growing, in California, 39
HEFBURN, JOSEPH SAMUEL: A critical study of the natural changes occurring in fats and oils, 365, 421
HIXON, HIRAM W.: Earthquakes in the light of the new seismology (correspondence), 227

Impact, The theory of, and its applications (Tiemann), 235, 336
India's mica industry, 39

Jamaica, Mineral wealth of (Outerbridge), 457

KNUDSEN, A. A.: The corrosion of metals underground by electrolysis, 132

LANDIS, EDWARD H.: Some of the laws concerning voltaic cells, 399
Lithographic stone industry, 301
Log rules, Maine commission reports, 216

MCALLISTER, A. S.: Improvement of power-factor and commutation conditions in single-phase series motors, 40
Maine, Peat deposits of, 199
Metals, Conservation of the, 185
Metals, Corrosion of, underground by electrolysis (Knudsen), 132
Mixteca country in the state of Oaxaca, Mexico (Birkinbine), 200
Motors, Single-phase series, Improvement of power-factor and commutation conditions in (McAllister), 40

Naval stores industry, Government studies, 106
Naval stores production in 1908 and 1907, reported, 217
Navigating the air (Post), 477
Newfoundland, Mineral wealth of (Outerbridge), 457
Newton's law and the cause of gravitation (Ely), 121

Obituary notice, Persifor Frazer, 75
OUTERBRIDGE, A. E., JR.: The mineral wealth of the islands of Newfoundland and Jamaica, 457

Panama canal, Present aspects of the, from the tourist's point of view (Tatham), 186
Peat as fuel, 197
Philadelphia district, geology of the, 18
Pig iron, Value of production of in 1908, 420
Pines, Southern, The effect of boxing or bleeding, 79
POST, AUGUSTUS: Navigating the air, 477
Power-house economics in Baltimore (Foster), 315
Proteins, Recent progress in the chemistry of the (Bradbury), 85
Publications received, 83, 314

RICE, CHARLES D.: Evolution in design, manufacture and uses of typewriting machines, 385
Road surfaces, modern, The development of (Fulweiler), 155, 260
Roadways, Rubber asphalt, 129

Scrub pine furnishes wood pulp material, 184

SEELY, LESLIE B.: Some problems in forestry, 1
Slate, Production of, in 1908, 198
Smoke nuisance, The, 120
Southern appalachian region, Ores and minerals of the, 115
STOUGHTON, BRADLEY: recent progress in open hearth steel practice, 470
Steel, open hearth practice, Recent progress in (Stoughton), 470
Submarine, Develop the (Balch), 108

TATHAM, WILLIAM: The present aspects of the Panama canal from the tourist's point of view, 186
THAYER, RUSSELL, The dirigible balloon with gyroscope control, 19
TIEMANN, HARRY D.: The theory of impact and its applications, 235, 336
Timber seasoning and wood preservation, 215
Titanium, 478
TRACY, MARTHA: Analysis of some Fairmount Park waters, 116
Typewriting machines, evolution in design, manufacture and uses of (Rice), 385

Vernaz milling files (Franklin Institute report), 219
Voltaic cells, Some of the laws concerning (Landis), 399

Waters, Fairmount Park, Analysis of some (Tracy), 116
Wood preservation, Timber seasoning and, 215
Wood preservatives, Bark effects, penetration of, 395
Wood preservatives, Increase in use of, indicates progress in forest conservation, 107
Wood preservatives used in 1908, 119